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Laser Cooling and Ion Beam Diagnosis of Relativistic Ions in a Storage Ring

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Relativistic Ions in a Storage Ring
Diagnosis of
Laser Cooling and Ion Beam

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Abstract

1878

During the winter and spring of the following year, I had the opportunity to visit the different parts of the country and to make some observations on the state of the country and the people. I found the country to be very fertile and the people to be very industrious. The climate was very pleasant and the scenery very beautiful. I was very much interested in the different parts of the country and the people and I was very much pleased to find that the country was so fertile and the people so industrious.

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Meiner lieben, geduldigen Frau,
die mich großartig unterstützt hat,
und unseren beiden Lütten
Friederike und Nils

Abstract

Particle accelerator and storage ring technology has reached an advanced state, so that different heavy ion storage rings are coming into operation by now, capable of storing even fully stripped ions up to U^{92+} . The main purposes of these machines are the accumulation of ions and the ability of improving the beam quality, that is the phase space density of the stored beams. This beam cooling is done successfully by the well established stochastic and electron cooling techniques. A new cooling method, the laser cooling, is taken over from atomic beam and ion trap experiments, where it has yielded extremely low temperatures of atomic samples.

As a candidate at storage rings ${}^7\text{Li}^+$ ions are stored in the Heidelberg TSR at 13.3 MeV. The ion beam properties of the metastable fraction like momentum spread, storage time and the influence of residual gas scattering are investigated by colinear laser spectroscopy in the experimental section of the TSR. The results are in agreement with the data from Schottky noise measurements, which is a standard diagnostic tool at storage rings. An optical pumping experiment using two dye laser systems yields information about ion kinematics and velocity mixing processes in the ring. Lifetimes in the order of 100 ms for velocity classes marked in this way show that laser cooling can be applied to the stored ${}^7\text{Li}^+$ beam.

In an experimental situation of two strong counterpropagating laser beams, both tuned near resonance, a dramatic reduction of the ion beam momentum spread is observed. With a special geometrical control of laser and ion beam the longitudinal beam temperature is reduced from 260 K to at least 3 K with very high collection efficiency. Schottky noise diagnosis of the cooled ion beam shows a strong enhancement of the cooled peak. This is interpreted as a collective phenomenon within the stored ion beam, representing a one-dimensional plasma, and confirms the strong cooling capability of resonant laser radiation in a storage ring.

Zusammenfassung

Die Teilchenbeschleuniger- und Speicherring-Technologie hat heute ein so hohes Niveau erreicht, daß zur Zeit mehrere Speicherringe für schwere Ionen gebaut werden. Verschiedenste Ionen bis hin zum U^{92+} können gespeichert werden. Zwei große Vorteile dieser Maschinen sind die Akkumulation von Ionen und die Möglichkeit, die Qualität des gespeicherten Ionenstrahls, die Phasenraumdichte, dramatisch zu verbessern. Diese Strahlkühlung wird bereits erfolgreich durch stochastische und Elektronenkühlung erreicht. Die neue Methode der Laserkühlung wird von Atomstrahl- und Fallenexperimenten übernommen, wo sie zu extrem niedrigen Temperaturen der atomaren Ensembles geführt hat.

Als ein Kandidat am Speicherring werden ${}^7\text{Li}^+$ Ionen im Heidelberger TSR bei 13.3 MeV gespeichert. Die Eigenschaften des metastabilen Anteils des Ionenstrahls werden durch kollineare Laserspektroskopie in der Experimentierstrecke des TSR untersucht, so z. B. die Impulsunschärfe, die Speicherzeit oder der Einfluß von Streuung am Restgas. Die Ergebnisse sind in Übereinstimmung mit Werten, die aus Messungen des Schottkyrauschens des Ionenstrahls gewonnen werden. Diese Methode ist ein Standard-Diagnoseverfahren an Speicherringen. Die detaillierte Bewegung der Ionen im Ring wird durch ein Experiment untersucht, das optisches Pumpen anwendet. Die Zeitskala für das Mischen von Geschwindigkeitsklassen im TSR kann so zu 100 ms bestimmt werden. Diese Ergebnisse der Strahldiagnose zeigen deutlich, daß die Laserkühlung auf gespeicherte ${}^7\text{Li}^+$ Ionen angewendet werden kann.

Im Experiment werden den Ionen zwei starke gegenläufige Laserstrahlen überlagert, die beide nahe der Resonanz stehen. Der Kühlzyklus führt zu einer drastischen Verringerung der Impulsbreite des Ionenstrahls. Bei spezieller Kontrolle des geometrischen Überlapps von Laser- und Ionenstrahl kann die longitudinale Temperatur der Ionen von 260 K mit sehr großer Effizienz auf weniger als 3 K reduziert werden. Im Schottkyspektrum des lasergekühlten Strahls ist die Verteilung der verschobenen und gekühlten Ionen im Verhältnis zu den ungekühlten Grundzustandsionen stark überhöht. Dieser Effekt wird als kollektives Phänomen in dem gespeicherten Ionenstrahl interpretiert. Als eindimensionales Plasma kann der Strahl zu Schwingungen angeregt werden. Daß diese Anregung durch resonantes Laserlicht erfolgen kann, macht die Effektivität von Laserwechselwirkungen im Speicherring deutlich.

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Chapter 1

Introduction

The 1989 noble prize in physics is dedicated to physicists working on electromagnetic trapping of particles and using them for high precision experiments. The Paul trap [Paul58] is capable of confining elementary particles as well as heavier ions to a small volume at a vanishing mean velocity in the laboratory frame. As a pioneer in precision experiments on single electrons stored and kept at rest in this way, Dehmelt measured the anomaly of the electronic g -factor to 10^{-11} [Wine73, Gabr85].

Particle traps with the apparently different feature of high mean velocity are particle storage rings, having been developed and operated for several years in high energy physics [Rubb85]. Here only a two dimensional confinement is established and the particles are allowed for free movement along the central orbit of the machine. For complex high energy experiments like the Z^0 -production in $p\bar{p}$ collisions at the CERN SPS, the cooling of stored particle beams is necessary. In order to collect up to 10^{11} antiprotons in the accumulator ring (AA), stochastic cooling is applied, increasing the phase space density of the beam by a factor of 10^9 [Meer85]. Another possibility of beam cooling, the electron cooling technique, has first been developed at NAP-M in Novosibirsk [Deme80] and has then also been pursued by CERN with the LEAR antiproton storage ring [Poth89].

In recent years, advanced accelerator technology made modifications of these elementary particle storage rings possible, so that by now different machines are coming into operation, which can store all kinds of ions up to U^{92+} (at the ESR in Darmstadt¹) at relativistic energies. A new field of atomic and nuclear physics is opened by these heavy ion storage rings, treated in more detail in chapter 2. Ions with at least one electron left, possess a detailed inner structure, which yields possibilities for precision spectroscopy as well as for a new technique of beam cooling by laser radiation. Laser cooling has been

¹Experimental storage ring at Gesellschaft für Schwerionenforschung in Darmstadt, FRG

applied very successfully to atomic beams. First experiments have been performed on laser cooling of free sodium atoms in a thermal beam [Andr81, Phil82] and the potential of this kind of velocity control has been demonstrated for various atomic species by now.

This technique can be carried over to ions in a storage ring. Besides the mean velocity of the ionic sample, the localisation of the ions is another major difference to conventional ion traps. In a trap the ion cloud is confined to a volume of about 0.1 cm^3 , while in a storage ring the ions are distributed in a toroid of at least $2 \cdot 10^3 \text{ cm}^3$. In this sense the control of the ion's position in a trap is equivalent to velocity control in a storage ring. Recent experiments in ion traps have shown, that laser cooling can yield this control very effectively [Died87a] and our experiment demonstrates, that this is also true for ion storage rings, where cooling means to compress the ion's velocity distribution towards monochromaticity. In both cases cooling is of fundamental importance for precision experiments. Exceeding stochastic cooling and electron cooling schemes, which operate successfully for high temperature and transverse motion, theoretically laser cooling may produce longitudinal temperatures T in the microkelvin regime, which is still an utopia for the cooling of solid samples. Recent theoretical and experimental studies for thermal atomic beams have shown that this limit can even be drastically lowered [Aspe88]. Optical and radiofrequency precision spectroscopy on ions in a trap [Neuh80, Blat82] have demonstrated the high performance of experiments on stored ions. The ionic sample has been cooled down even to the Lamb-Dicke regime, where the kinetic energy of an ion is below the first oscillation band of the harmonic trap potential [Neuh78, Died88]. New physical phenomena like clustering and crystallization have been observed on laser cooled ions in a trap [Died87a, Gilb88] and also in storage rings, the onset of ordered structures and phase transitions in the stored ion beam may come to the scope of experimental feasibility [Rahm86, Habs86, Died87a, Wine87].

In the frame of this work, laser spectroscopic experiments have been performed on metastable ${}^7\text{Li}^+$ ions, stored at kinetic energies of 9.12 and 13.3 MeV respectively in the Heidelberg TSR ². Different important features are pointed out.

Ion beam diagnosis can be performed with very high accuracy as discussed in chapter 4. The local velocity is measured and velocity changes due to energy losses by residual gas scattering or due to photon momentum transfer are detected. The lifetime of the metastable fraction of the beam is measured and the momentum spread can be deduced

²Test storage ring at Max Planck – Institut für Kernphysik in Heidelberg, FRG

from the fluorescence signal, giving a measure for cooling and heating processes.

Optical pumping signals show, that these diagnostical features are of fundamental importance for high precision experiments in a storage ring (section 4.3). The observed Lamb dips are broadened due to the specific ion kinematics in the storage ring, which is briefly outlined. The lifetime of particular velocity classes is measured and discussed.

The last part of this work reports the improvements of ion beam properties by laser cooling of the stored beam. The longitudinal ion velocity distribution is compressed very effectively, and it can be proved, that almost 100% of the metastable ions are accumulated in a narrow velocity range. Taking electronic Schottky noise spectra of the cooled ion beam gives evidence for the onset of collective phenomena in the cold ensemble of stored ions.

Chapter 2

Ion Storage Rings

In order to give an overview of the many activities in the new field of ion storage ring physics I shall report here the atomic physics programme at the ESR with major respect to laser experiments. Then the TSR is presented in its layout and main features of operation.

2.1 The ESR Atomic Physics Program

The ESR in Darmstadt is the most ambitious project of the various ion storage rings in operation or under construction. Beams of even fully stripped ions will be stored at highest possible phase space densities. Maximum kinetic energies of 1.3 GeV/u for U^{92+} ions are projected and stochastic as well as electron cooling devices are implemented in the ring. They allow to reduce the beam emittance to $\varepsilon = 0.1 \pi \text{ mm mrad}$ and the momentum spread down to $\Delta p/p \leq 10^{-5}$ [Fran86, Fran87, Armb88, Kien89].

Many interesting proposals have been made for nuclear physics experiments at the ESR like fusion fragment separation and accumulation [Armb88]. Cooling the radioactive beams to high phase space density will cause dramatic progress in collision experiments or accelerator mass spectroscopy. In the atomic physics field large cross sections exist for photon ion interaction and for inelastic collisions between ions and slow electrons from the electron cooling beam¹. The different recombination processes in such slow collisions are summarized in table 2.1 and the well known time reversed processes are indicated [Beye88]. The dielectronic recombination, an important process in hot plasmas, has been studied experimentally under clean conditions at the TSR [Kilg90]. The REC may be enhanced by resonant laser radiation [Neum83, LaGa88], although experimentally there are limitations due to available laser wavelengths and low rate coefficients for accessible

¹These electrons are slow with respect to the moving frame of the ions. For electron cooling the electron velocity is tuned to match the ion velocity.

Table 2.1: Ion – electron recombination processes

process	reaction
radiative electron capture (REC)	$A^{Z+} + e^- \longrightarrow A^{(Z-1)+} + h\nu$
threebody recombination (TR)	$A^{Z+} + e^- + e^- \longrightarrow A^{(Z-1)+} + e^- + E_{kin}$
dielectronic recombination (DR)	$A^{q+} + e^- \longrightarrow A^{(q-1)+**} \longrightarrow A^{(q-1)+*} + h\nu$
	time reversed process
REC	photoionization
TR	electron impact ionization
DR	Auger process

electronic states.

Direct laser spectroscopy of stored ions e.g. aims at fundamental QED studies in high Z ions. Fine structure and hyperfine structure splittings scale with Z^3 and there are several multiply charged ion species, where the corresponding M1 transition frequencies lie in the optical region [Kühl87a]. Laser excitation of these forbidden transitions will be applied as a high precision method and like in many recent experiments on ions stored in ion traps [Boll85, Dehm87] highest optical resolution will be achievable.

Due to the large first order Doppler shift the possibilities for laser spectroscopy will be extended to the infra-red or far uv region in rings. The 154 nm ground state transition in the lithiumlike C^{3+} ion e.g. will be shifted by a factor of 2.09 at 264 MeV/u kinetic energy in the ESR.

A unique application of the high relativistic velocity of the stored ions lies in the field of testing the theory of special relativity by measuring the one way speed of light [Robe49, Mans77, Kaiv85]. ${}^7Li^+$ e.g. can be stored in the ESR at velocities of up to 40% of the speed of light. In a saturation spectroscopy experiment on the moving ions, the frequencies of the co- and counterpropagating laser beams have to be measured to high accuracy. Together with the very well known spectroscopy of the ${}^7Li^+$ ion at rest, the precision in measuring the quadratic Doppler effect can be improved substantially [Hube87]. Extending these measurements over days or months will check the siderial constancy of the speed of light, a fundamental question in astrophysics [Riis88].

This brief overview is far from being complete, but shows the possibilities in this new field of research. Although the ESR is just starting operation, first experiments in many different fields have already been performed at the TSR, which has been running since 1988 [MPI88, MPI89].

2.2 TSR Layout and Operation

The heavy ion storage ring of the Max-Planck-Institut für Kernphysik in Heidelberg (TSR) has been constructed with a supersymmetry of two and with a high geometrical and momentum acceptance $\frac{\delta p}{p} \leq \pm 3 \cdot 10^{-2}$ [Arno86, Baum87, Habs89]. Table 2.2 gives the maximum kinetic energies, at which some specific ions can be stored in the TSR at its maximum magnetic rigidity of 1.7 Tm. Lithium and beryllium ions are the most rigid

Table 2.2: Maximum kinetic energies at TSR

ion	$p/\frac{\text{MeV}}{c}$	$E_{kin}/\frac{\text{MeV}}{u}$
p	510	130
$^{12}\text{C}^{6+}$	3060	34.3
$^7\text{Li}^+$	510	2.84
$^9\text{Be}^+$	510	1.72

ones stored in the TSR up to now. In figure 2.1 the lattice layout of the TSR is shown. Devices for electron cooling and rf-bunching are implemented in the acromatic sections of the ring. In the experimental section opposite to the injection, specific user setups can be installed.

The ion storage ring is set up with the EN Tandem and the post accelerator. In a pulsed high current sputtering ion source LiH^- ions are produced and pre-accelerated. In order to obtain kinetic energies of 9.12 MeV or 13.34 MeV, which have both been used for the experiments described here, the ions are passed through the stripper of the Tandem at a kinetic energy of 4.66 or 6.76 MeV respectively. The Li^+ -fraction is produced partially in the $^1\text{S}_0$ ground and $^3\text{S}_1$ metastable state. The $^7\text{Li}^+$ bunches are fed into the TSR in multiturn injection mode. During the injection cycle the whole geometrical acceptance of the beam tube is filled with about 40 bunches to 2 - 8 μA ion beam current and they spread out to form a caustic beam after several microseconds. It is possible to accumulate 20 times this intensity by rf-stacking, which makes use of the high momentum acceptance of the TSR. In addition to the multiturn mode it fills up all the momentum phase space, but since the rf-stacking increases the momentum spread of the stored ion beam drastically, we did not use this technique. That means a maximum of $2 \cdot 10^8$ $^7\text{Li}^+$ ions circulate in the ring.

The storage time of the beam is limited by collisions with residual gas molecules leading to a particle loss due to ionization, scattering or electron capture and thus it

laser beam
(contralinear)

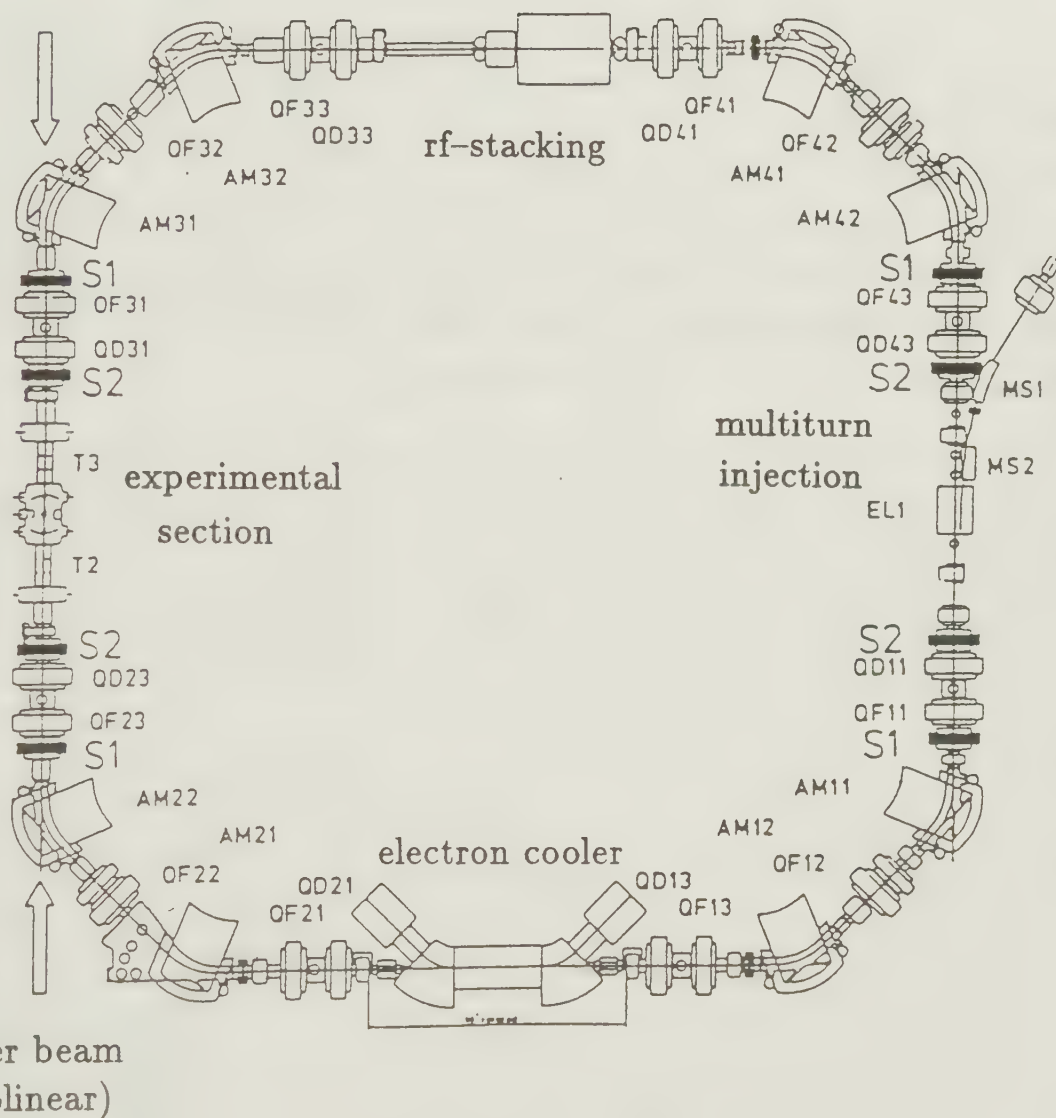


Figure 2.1: Lattice layout of the TSR. Besides the main dipole magnets for bending of the ion beam quadrupole and sextupole magnets are necessary for focussing and higher order corrections. The four straight sections of the ring are used for injection, electron cooling, applying radiofrequency to the beam and for implementation of experimental setups.

The two laser systems are located in the basement below the experimental hall, and the laser beams are guided for about 18 m into the experimental section.

depends strongly on the composition of the residual gas. The contribution of each of these processes to the total storage time τ can be estimated theoretically [Hint89, Habs89]. τ may be dominated by one of these three loss processes at specific operating conditions of the TSR. In the case of ${}^7\text{Li}^+$ the stripping of a second electron is the most probable, especially for the excited metastable 3S_1 electron. The maximum total beam lifetime for ${}^7\text{Li}^+$ observed at the TSR is 24 s at an average pressure of 10^{-10} mbar.

For relatively weak beams the beam position and lifetime can be measured in the bunched beam mode. Conditions are considerably better for intense beams of higher charged ions. A ${}^{12}\text{C}^{6+}$ beam e.g. has been stored at 73.3 MeV total kinetic energy with intensities of 18 mA for 1 hour without and for 5 hours with electron cooling [Habs89, Stec90]. At our particular experimental conditions, however, we are at the lower threshold for electronic ion beam diagnosis via Fourier analysis of the longitudinal Schottky noise of the ion beam.

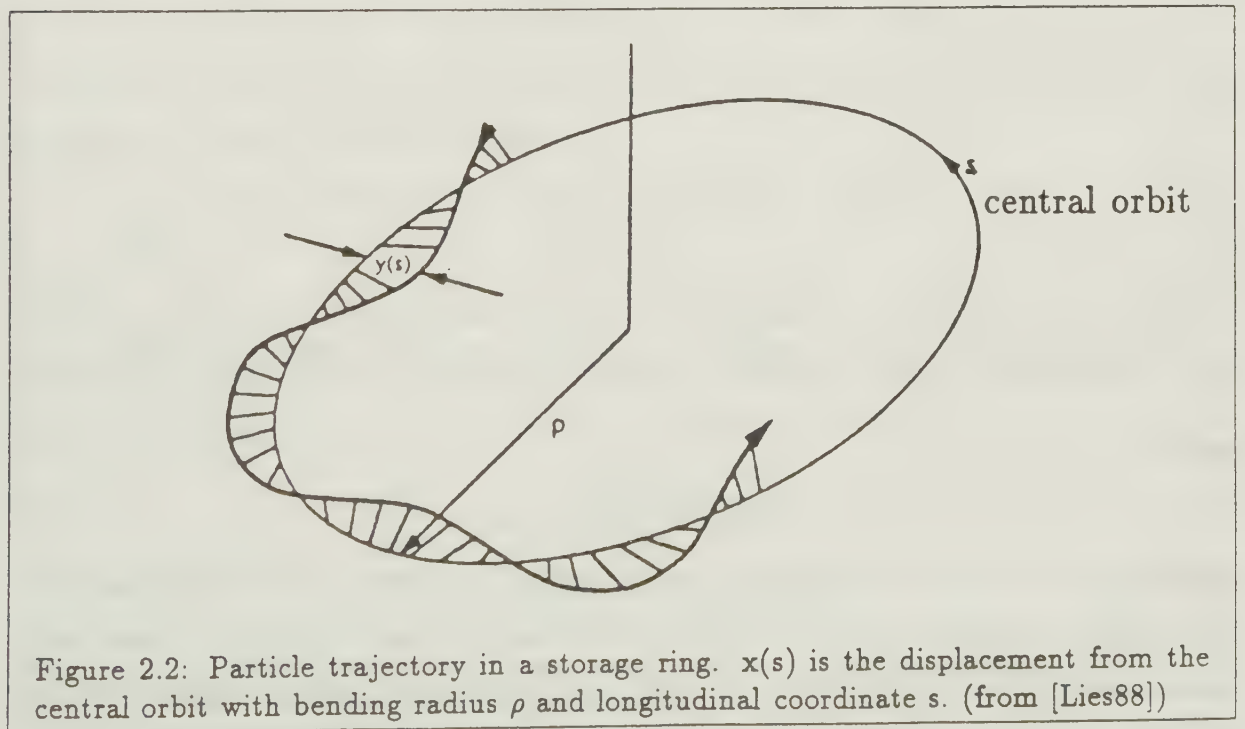
2.3 Storage Ring Kinematics

It is necessary for a detailed understanding of the influence of laser interaction and cooling on the ion motion to give a description of the ion kinematics in a storage ring. These machines basically consist of different types of ion optical elements, guiding and focusing the beam. Their influence can be expressed in the well known matrix formalism [Sept67], but in storage ring physics a more general differential equation description is used.

In contrast to their free motion along the central orbit of the storage ring, the ions are confined in both other directions perpendicular to this z -axis. This confinement is established by the strong focussing elements of the ring lattice and allows for harmonic oscillations of the ions around the 'sollbahn' [Bry85]. These transverse oscillations are described by a differential equation of Hill's type.

$$\frac{d^2 x}{ds^2} + K(s)x = \frac{1}{\rho} \frac{\Delta p}{p_0} \quad (2.1)$$

where x is the displacement from the central orbit in the vertical or horizontal plane, s is the longitudinal coordinate, ρ is the bending radius in a dipole, $K(s)$ is the focussing strength and $\Delta p/p_0$ is the relative deviation from the ideal central orbit momentum p_0 (see fig. 2.2). This equation yields the position and momentum of each particular ion in



the ring, but regarding an ion beam with typically $10^8 - 10^{10}$ stored ions quantities, like

the emittance ε , or the beam envelope $E(s)$, are of interest. The displacement x of an ion as given by solution of 2.1 for zero momentum deviation ($\Delta p = 0$), increases with the emittance ε and with the betafunction $\beta_s(s)$ of the storage ring

$$x(s) = \sqrt{\varepsilon \beta_s(s)} \cdot \cos(\psi(s) - \delta) \quad , \quad (2.2)$$

while the s dependence includes a variable phase δ and the betatron phase $\psi(s) = 2\pi \cdot \int \frac{ds}{\beta_s(s)}$. The betafunction of a storage ring is determined by the lattice layout; that is the arrangement of magnets along s , and the operating parameters. The specific situation for measurements described here may be seen in fig. 2.3. The tune Q of the storage ring is

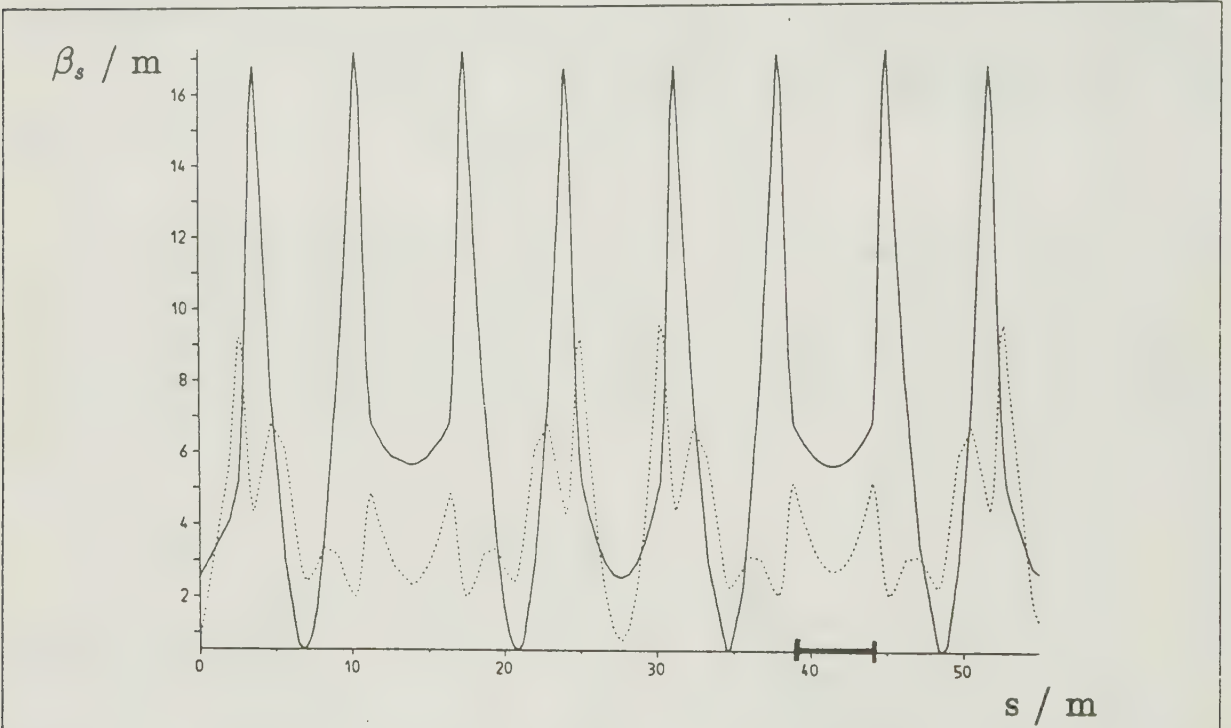


Figure 2.3: Horizontal (—) and vertical (···) betafunctions of the TSR for ring settings used in experiments described here. The laser cooling section is marked.

given by

$$Q = \oint \frac{ds}{\beta_s(s)} \quad . \quad (2.3)$$

Physically Q is the number of betatron oscillations in one revolution, and it is greater than 1 for strong focussing machines. The value of Q has to be nonrational in order to avoid resonances with the structure of the storage ring lattice. If Q meets a rational number, distortions of the trajectory of a particle will add up and the beam is lost quickly. The beam envelope $E(s)$ is just the maximum displacement for any ion and arbitrary value of

the betatron phase

$$E(s) = x_{\max}(s) = \sqrt{\varepsilon \beta_s(s)} \quad . \quad (2.4)$$

With an adopted value for the emittance $\varepsilon = 60\pi \text{ mm mrad}$, $\beta_{s,x} = 5.1 \text{ m}$ and $\beta_{s,y} = 1.9 \text{ m}$, we calculate a beam size in the experimental section of the TSR of 3.6 cm in horizontal and 2.2 cm in vertical direction. The divergence $x' = \frac{dx}{ds}$ is given by

$$x'(s) = \sqrt{\frac{\varepsilon}{\beta_s(s)}} \left(\sin(\psi(s) - \delta) + \frac{1}{2} \beta'_s \cos(\psi(s) - \delta) \right) \quad (2.5)$$

determined by the local parameters β_s and $\beta'_s = \frac{d\beta_s}{ds}$. For the transverse velocity at a given point s we get

$$v_x(s) = \frac{dx}{ds} \frac{ds}{dt} \quad (2.6)$$

$$= \left(\sqrt{\frac{\varepsilon}{\beta_s(s)}} \left(\sin(\psi(s) - \delta) + \frac{1}{2} \beta'_s(s) \cos(\psi(s) - \delta) \right) \right) \cdot v_0 \quad (2.7)$$

where we introduced the mean ion velocity $v_0 = \frac{ds}{dt} = \frac{L\omega_0}{2\pi}$ with L the ring circumference and ω_0 the revolution frequency. Similiar equations are valid for the y direction. The constant total ion velocity is given by

$$v_0 = \sqrt{v_x^2 + v_y^2 + v_z^2} \quad , \quad (2.8)$$

but as in laser measurements we are sensitive only to v_z , we calculate the modulation of v_z induced by the transverse oscillations.

$$v_z = \sqrt{v_0^2 - v_x^2 - v_y^2} \quad (2.9)$$

$$\frac{v_z}{v_0} \approx 1 - \frac{1}{2} \frac{v_x^2 + v_y^2}{v_0^2} = 1 - \frac{\delta v_z}{v_0} \quad . \quad (2.10)$$

Averaged over many turns, that is over the betatron phase $\psi(s)$, the modulation of v_z at a given point s has its maximum value for one transverse motion at

$$\frac{\delta v_z}{v_0} = \frac{1}{2} \frac{\varepsilon}{\beta_s(s)} \left(1 + \frac{1}{4} \beta_s'^2(s) \right) \quad (2.11)$$

$$= \frac{1}{2} \frac{E^2(s)}{\beta_s^2(s)} \left(1 + \frac{1}{4} \beta_s'^2(s) \right) \quad . \quad (2.12)$$

In the middle of the experimental section, where $\beta'_s = 0$, the maximum relative transverse velocities $\frac{v_x}{v_0}$ and $\frac{v_y}{v_0}$ are in the order of a few times 10^{-3} , resulting in a total variation $\frac{\delta v_x}{v_0} = 1.6 \cdot 10^{-5}$. Off this center $\frac{\delta v_x}{v_0}$ gets larger according to the growing derivative $\beta'_s(s)$. Due to the Doppler effect this variation of the longitudinal velocity of an ion also changes

the Doppler shifted resonance frequency (see sec. 3.1). This inherent line broadening can be minimized only by a reduction of the amplitudes of betatron oscillations, that is either transverse beam cooling or simply collimating the ion beam by mechanical apertures.

The beam injected into the storage ring has a momentum spread defined by the preceding accelerator. In the ring, ions with momenta differing from p_0 will travel on different paths compared to the central orbit. The closed orbit for $\Delta p = (p - p_0) \neq 0$ shows a horizontal displacement $\Delta x = D(s) \frac{\Delta p}{p_0}$, where $D(s)$ is the dispersion function of the ring. For an upper limit of the momentum spread of the ${}^7\text{Li}^+$ beam of 10^{-4} this displacement is with 0.16 mm much smaller than the horizontal size of the total beam and also well below the typical size of the applied laser beams. Nevertheless, as a consequence, the revolution frequency of this particle differs from ω_0 by $\Delta\omega$.

$$\frac{\Delta\omega}{\omega_0} = \left(\frac{1}{\gamma^2} - \frac{1}{\gamma_{tr}^2} \right) \frac{\Delta p}{p_0} \quad (2.13)$$

γ_{tr} is the so-called 'transition gamma', which in most cases is approximately equal to the horizontal tune Q_h . By this relation the distribution of revolution frequencies of the stored ions is a measure of the beam momentum spread.

Chapter 3

Laser Spectroscopy in a Storage Ring

The application of laser spectroscopy to fast ions in a storage ring demands detailed relativistic treatment of the ion motion and the transformation between the laboratory frame and the rest frame of the stored ion ensemble.

3.1 Relativistic Kinematics

Ions at relativistic velocities are described by their 4-vectors $(ct, \vec{x})$ and $(\frac{E}{c}, \vec{p})$. The Lorentz transformation of an arbitrary 4-vector $(A, \vec{A})$ from the system S to the frame S' moving along the z -axis at the velocity $\beta = \frac{v}{c}$ is given by

$$A'_0 = \gamma(A_0 - \beta A_z) \quad (3.1)$$

$$A'_{x,y} = A_{x,y} \quad (3.2)$$

$$A'_z = \gamma(A_z - \beta A_0) \quad (3.3)$$

$\gamma = (1 - \beta^2)^{-\frac{1}{2}}$ denotes the usual relativistic factor. From these equations the known formulas for the transformation of length, time and velocity follow directly.

For ions of rest mass $E_0 = m_0 c^2$, accelerated to a kinetic energy E_{kin} and a momentum p , the velocity β is connected to these quantities via

$$\beta = \sqrt{1 - \left(\frac{1}{1 + \frac{E_{kin}}{E_0}} \right)^2} \quad (3.4)$$

In order to excite an internal optical transition of the moving ion with energy spacing $E_{h\nu} = h\nu_0$ by laser radiation, the frequency shift due to the Doppler effect has to be taken into account. A plane light wave with angular frequency $\omega = 2\pi\nu$ and propagation vector

$\vec{k}$ transforms its 4-vector $(\frac{\omega}{c}, \vec{k})$ and one finds in the moving frame

$$\nu' \equiv \nu_{\pm} = \gamma \nu_0 (1 \pm \beta \cos \Theta) \quad (3.5)$$

for the relativistic Doppler effect. The angle Θ transforms like

$$\tan \Theta' = \frac{\sin \Theta}{\gamma (\cos \Theta - \beta)} \quad (3.6)$$

ν_0 is the optical transition frequency in the ion's rest frame and Θ the angle between the ion's velocity vector and the laser beam direction $\vec{k}$. Knowing the angle Θ or approximating $\cos \Theta \approx 1$ for small Θ , the ion velocity may be determined by measuring the laser resonance frequency ν_+ or ν_- with the additional knowledge of ν_0 . In the case of perfect colinear geometry ($\Theta = 0$) we find from equation 3.5

$$\beta = \frac{\frac{\nu_{\pm}^2}{\nu_0^2} \mp 1}{\frac{\nu_{\pm}^2}{\nu_0^2} \pm 1} \quad (3.7)$$

A variation $\delta\beta$ of this velocity or δE_{kin} of the ion's kinetic energy results in a corresponding change of the Doppler shifted frequency.

$$\delta \nu_{\pm} = \nu_{\pm} \gamma^2 \delta \beta = \nu_0 (1 \pm \beta) \gamma^3 \delta \beta \quad (3.8)$$

$$\delta \nu_{\pm} = \nu_{\pm} \frac{1}{\beta \gamma} \frac{\delta E_{kin}}{m_0 c^2} = \nu_0 \frac{1 \pm \beta}{\beta} \frac{\delta E_{kin}}{m_0 c^2} \quad (3.9)$$

This is the principle of the Doppler cooling technique, where the ions are kept in resonance while they are decelerated and accumulated in a narrow frequency range by a sweep of the cooling laser. A single absorbed photon of energy $E_{\gamma} = h\nu$ decelerates a ${}^7\text{Li}^+$ ion by $\Delta\beta = \gamma^{-3} \frac{E_{\gamma}}{E_0} = 3.5 \cdot 10^{-10}$ ($\Delta v = 10.3$ cm/sec), which results in a frequency shift of 180 kHz for counterpropagating geometry at $\beta = 0.064$.

The modulation of the longitudinal ion velocity due to the betatron oscillations, as calculated before (see sec. 2.3), results in a broadening $\delta\nu_B$ of optical resonances depending on the betatron amplitude or the ion beam diameter. Some numbers calculated for our specific settings in the middle of the experimental section ($\Delta s = 0$) and 1 m apart are given in the following table.

Table 3.1: Induced line broadening due to betatron oscillations

ion beam diameter	$\delta\nu_B (\Delta s = 0 \text{ m})$	$\delta\nu_B (\Delta s = 1 \text{ m})$
36 mm x 22 mm	584 MHz	612 MHz
22 mm	514 MHz	543 MHz
18 mm	345 MHz	364 MHz
10 mm	106 MHz	112 MHz
5 mm	27 MHz	28 MHz
1 mm	1.1 MHz	1.1 MHz

3.2 The ${}^7\text{Li}^+$ Ion as the Candidate

There are only few ion species with strong E1 transitions within the optical frequency range, which are suitable for laser cooling. The condition, that such a transition has to start from the ionic ground state or from a longlived metastable state and that it has to be a closed system, restricts the application of laser cooling mainly to the ion species listed in table 3.2¹. If one takes additional lasers into account for repumping the population of

Table 3.2: Ion species suitable for laser cooling

	${}^7\text{Li}^+$	${}^9\text{Be}^+$	${}^{24}\text{Mg}^+$
lower level	$2s\,{}^3S_1(F = 5/2)$	$2s\,{}^2S_{1/2}(F = 2)$	$3s\,{}^2S_{1/2}$
upper level	$2p\,{}^3P_2(F = 7/2)$	$2p\,{}^2P_{1/2}(F = 1)$	$3p\,{}^2P_{1/2}$
transition wavelength	548.5 nm	313 nm	280 nm
lifetime of lower level	59.8 s	stable	stable
lifetime of upper level	43 ns	8.7 ns	3.7 ns
Doppler limit ¹	89 μK	440 μK	1050 μK
recoil limit	4.5 μK	10.9 μK	5.1 μK

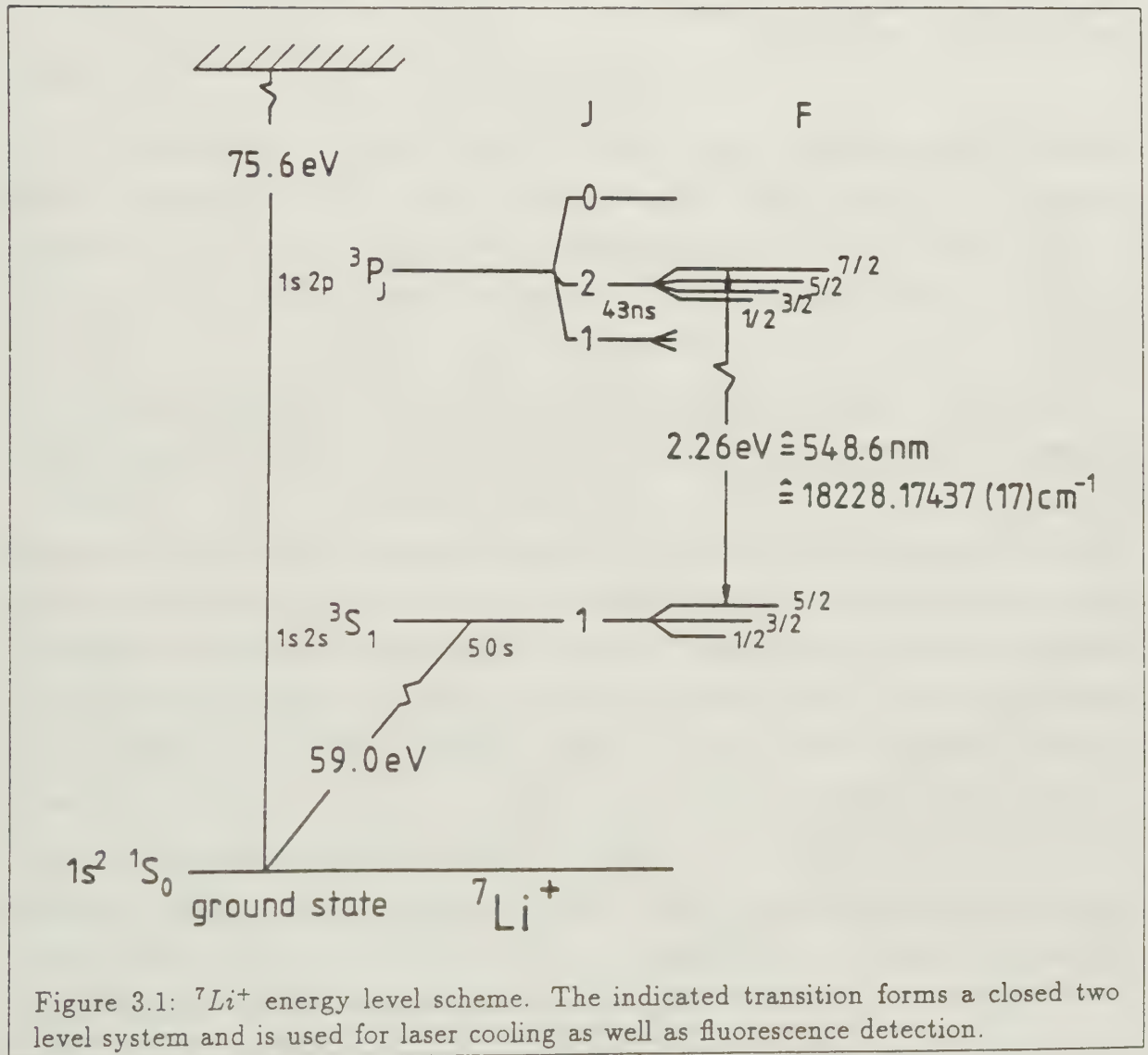
a possible third eigenstate of the ion, this list extends by heavier ions like ${}^{88}\text{Sr}^+$, ${}^{138}\text{Ba}^+$ or ${}^{198}\text{Hg}^+$ e.g. [Ertm84, Died89].

For laser spectroscopic experiments and cooling in an ion storage ring, the helium-like ${}^7\text{Li}^+$ ion satisfies all demands perfectly. Its 548.5 nm transition in the triplet system [Fan78, Baye79] starts from the triplet ground state with a radiative lifetime of 59.8 s [Knig80] (see figure 3.1). Both Doppler shifted transition frequencies, ν_+ and ν_- , lie in the optical region, even for extremely high ion velocities accessible at the ESR in Darmstadt. For the projected precision experiment using saturation spectroscopy on the moving ions with co- and counterpropagating laser beams [Hube87], ${}^7\text{Li}^+$ is the only possible ion to use. In addition it is useful in this sense, that the particular transition in the ${}^7\text{Li}^+$ ion at rest, is known to high accuracy by different theoretical and experimental investigations [Baye79, Riis86].

For the purpose of laser cooling it is of some convenience that the hyperfine splitting in the 3S_1 and 3P levels in ${}^7\text{Li}^+$ is very large and lies in the order of 10 – 20 GHz. This implies, that the $2s\,{}^3S_1(F = 5/2) \rightarrow 2p\,{}^3P_2(F = 7/2)$ transition represents a very clean two level system, which is needed for laser cooling. There is no considerable

¹for laser cooling limits see chapter 5

optical pumping to neighbouring hyperfine sub-levels and the spontaneous decay leads back to the $2s\ ^3S_1$ ground state of the triplet system. Due to singlet – triplet mixing via E1 and M2 transitions, only a 10^{-5} branching ratio exists to the $1s\ ^1S_0$ ionic ground state [Prio80]. This value has been measured on $^7\text{Li}^+$ ions in an rf quadrupole trap and is averaged over all $F=1/2 \dots 7/2$ sub-states. The $F=7/2$ state on its own does not show any mixing because there is no counterpart in the singlet system. The saturation intensity, needed to drive the cooling transition effectively, is $I_S = 8.8 \frac{\text{mW}}{\text{cm}^2}$ (see appendix A).



3.3 Stability of metastable ${}^7\text{Li}^+$ in the TSR

Besides the overall storage lifetime of the ion beam, in the case of ${}^7\text{Li}^+$, we have to be aware of some mechanisms, which might quench the metastable 3S_1 state in the storage ring. In section 2.2 we have already seen that the most probable process in a collision with residual gas molecules, leading to a particle loss, is the stripping of an electron. The cross-section σ_{el} for this process depends on the ionization energy of the ion, and thus the metastable fraction of the beam is expected to die out 2.1 times more quickly than the ground state ions [Niko65].

$$\sigma_{el} \propto \frac{1}{\sqrt{E_{ion}}} \quad (3.10)$$

If a metastable ion undergoes such a collision but is not ionized, it may, however, be de-excited to the 1S_0 ground state. By spectroscopical means this is equivalent to a loss of an 'active' ion, but it is obvious, that this process is neglectable compared to ionization.

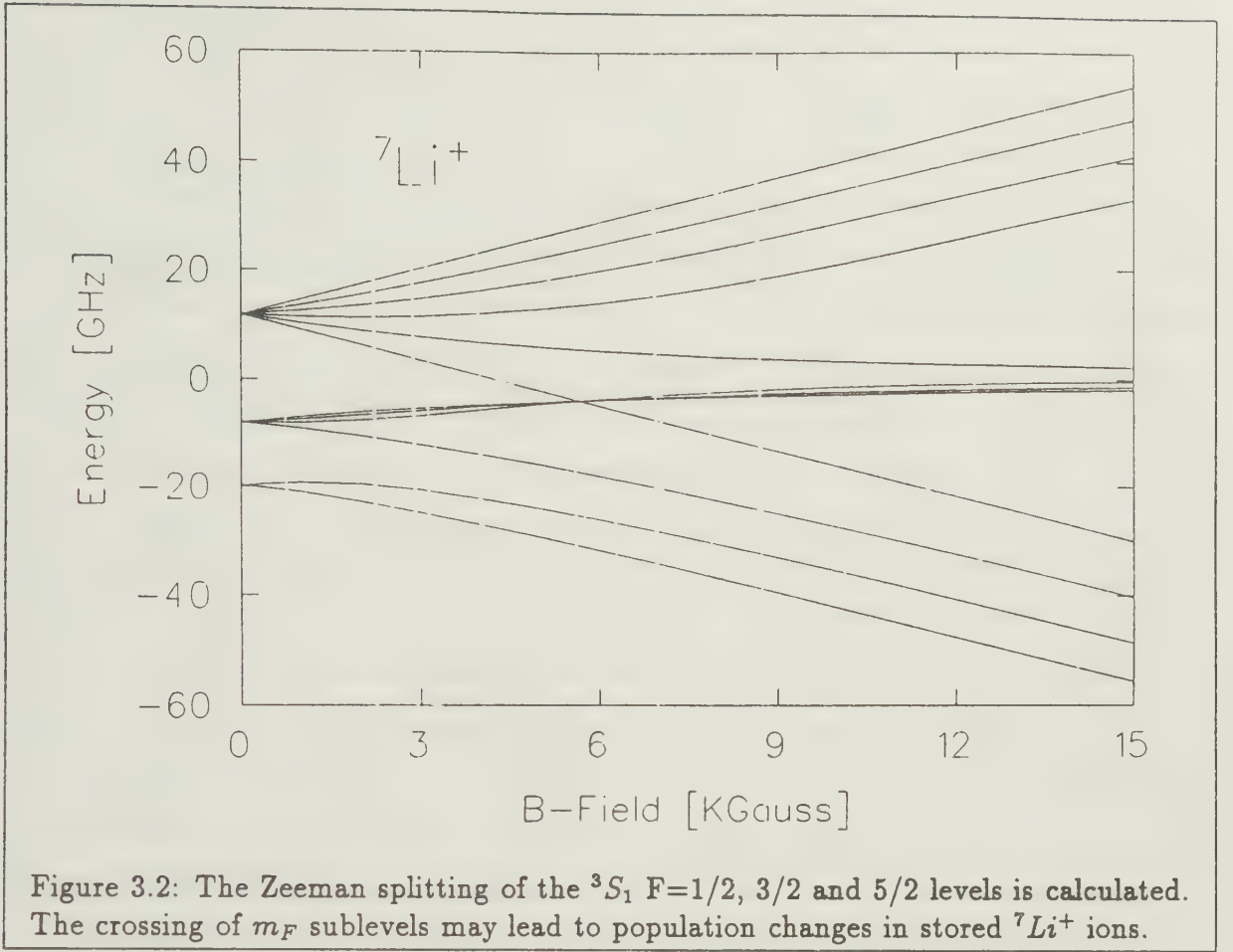
Further quenching mechanisms are introduced by the storage ring lattice. An ion moving in the TSR exhibits extreme electromagnetic field conditions. The dipole magnets, for example, produce a magnetic field gradient of about $\frac{1T}{10cm}$. Moving ions traversing such field gradients can be regarded as if irradiated by microwave radiation. A Fourier analysis of these fields for 13.3 MeV ${}^7\text{Li}^+$ ions, yields strong components at 200, 17 and 3.8 MHz. In the strong magnetic fields of the storage ring, the energy levels of the 3S_1 state are shifted drastically due to the Zeeman effect (see fig. 3.2). However, only the $m_F = -5/2$ sublevel of the $F = 5/2$ state crosses with the $F = 3/2$ sublevels. During this adiabatic process part of the $F = 5/2$ population might be lost, but the opposite way is also possible. So this effect of level mixing will lead to a partial inclusion of the $F = 3/2$ and $F = 1/2$ state ions to the procedure of laser cooling.

A third quenching mechanism, which does not only mix the F-states, but leads to a de-excitation to the ion's ground state, is due to the quadratic Stark effect induced by the motional electric field in the storage ring.

$$\vec{E} = \frac{\vec{v}}{c} \times \vec{B} \quad (3.11)$$

$$|\vec{E}| \leq 2.7 \cdot 10^7 \frac{V}{m} \quad (3.12)$$

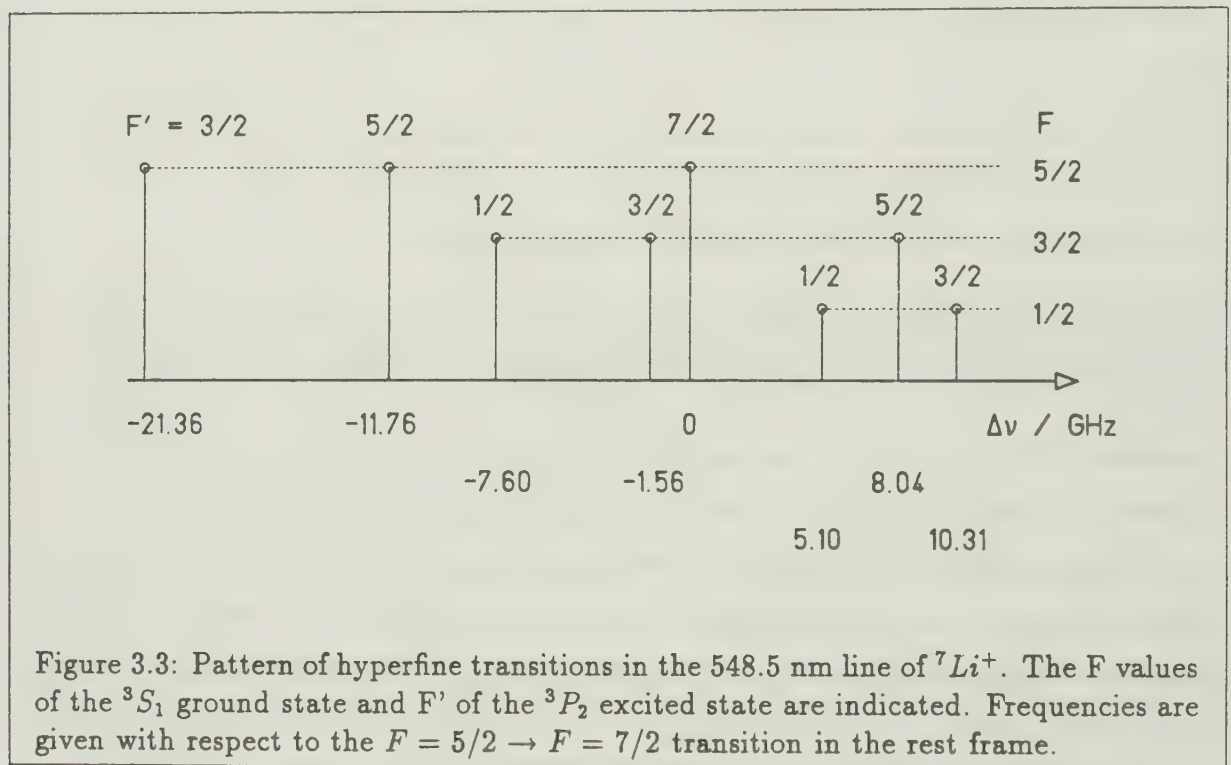
For this maximum value the wave function of the lower cooling level 2^3S_1 will contain a 10^{-4} admixture of the 2^3P_1 fine structure state. Via the branching of $\frac{1}{1200}$ due to LS-mixing from here to the 1S_0 ground state [Prio80], the decay probability of the 3S_1 state



is estimated to $4.6 \cdot 10^{-2}/s$. The associated lifetime of 22 s due to motional Stark effect mixing is of the same order as the radiative lifetime of this level (see chap. 3.2).

Regarding the laser excitation of the ions, the laser and ion beam are overlapped between two dipole magnets (see fig. 2.1). If the antiparallel laser is tuned to resonance for ions within the straight section of the ring, the ions see a red-shifted laser frequency when turning around the bend in the dipoles. During this time very few optical transitions to hyperfine substates other than the $2^3P(F=7/2)$ may be induced, causing such ions to be lost for the laser cooling cycle. The $F=5/2 - F'=5/2$ transition e.g. is resonant at a bending angle of $\alpha = 2.6 \cdot 10^{-2}$ for a time interval of about 2 ps every turn. This leads to a net optical pumping time of $5.4 \cdot 10^{-7}$ in a second and thus to a minimum lifetime (at saturation) of ≈ 200 ms due to such population mixing. Optical pumping may also occur by excitation of a neighbouring hyperfine transition due to the tail of the Lorentz shaped natural linewidth. The hyperfine pattern of the $^7Li^+$ spectrum in the 548.5 nm line is shown in figure 3.3. The frequency scale is given in the rest frame, and the nearest transition starting from the $F = 5/2$ level is suppressed by a factor of $(\frac{\delta\nu_{nat}}{11.78 \text{ GHz}})^2 \approx 10^{-7}$.

The contribution of the Gaussian profile at a Doppler width of 2 GHz is neglectable. Thus, saturating the $F=5/2 \rightarrow F'=7/2$ transition at a rate of $1.1 \cdot 10^7/s$, a transition to the $F'=5/2$ state will occur once a second. According to the branching of 1:1.4 from here to the $F=3/2$ and $F=5/2$ level, within two seconds the ion will be pumped to the $^3S_1(F=3/2)$ level. However, the effective interaction time of stored ions with the laser beam is reduced by different factors as discussed in section 5.3. Taking only the evident duty cycle $\eta = 0.09$ due to the circulation of the ions into account, the lifetime of the $F=5/2$ population associated with this off-resonance excitation is at least in the order of 20 s. The reverse $F=3/2 \rightarrow F'=5/2$ transition, which is excited slightly more frequently at a rate of about 0.2/s, partly repumps the $F=3/2$ population. In conclusion to these



various possibilities of losing an ion for the laser cooling cycle, the time for an ion to stay in the 3S_1 state can be estimated to be at least 20 s before it is lost to the singlet system. Optical pumping within the F substates, however, may occur on a shorter timescale as low as 0.2 s.

Chapter 4

Ion Beam Diagnosis

Laser fluorescence spectroscopy may give very precise information about ion beam properties of interest in a storage ring. A laser measurement is sensitive to the local velocity of an ion in the detection region and thus it is not sensitive to collective plasma waves e.g., or disturbed by high ion densities like the Schottky noise analysis.

Especially in the case of ${}^7\text{Li}^+$ it is of some advantage for laser diagnostics in a storage ring, that the rest frame transition frequency ν_0 of the system ${}^3S_1(F = 5/2) \rightarrow {}^3P_2(F = 7/2)$ is known to high accuracy by fast beam collinear laser spectroscopy [Rüs86].

$$\nu_0 = 18228.17437(17)\text{cm}^{-1} \quad (4.1)$$

4.1 Optical setup at the TSR

The laser equipment at the TSR is located in the basement of the experimental hall in a specially constructed laser hut. For solving the many different questions accessible to laser spectroscopy at the TSR, also quite different realizations of the optical setup have been used. In figure 4.1 two cw dye laser systems with the accompanying diagnostical equipment are shown, as they are used for the first experiments to be described. At 9.21 MeV ion energy, two single-mode cw dye laser systems are used, one operated with R6G and one with R110 or R6G dye respectively. The ring laser is tuned via a microcomputer and is set to 578.4 nm on the ${}^3S_1 \rightarrow {}^3P_2$ resonance in collinear counterpropagating geometry. The linear laser is set on fixed frequencies at 520.4 or 578.4 nm for co- or counterpropagating resonance. For the final laser cooling experiments, when the ion energy was raised to 13.34 MeV, this setup is modified by introducing a single-mode Ar^+ ion laser instead of the standing wave dye laser system (see 6.2). The optical frequencies are measured using iodine molecular absorption lines and a commercial wavemeter (Burleigh, model WA20).

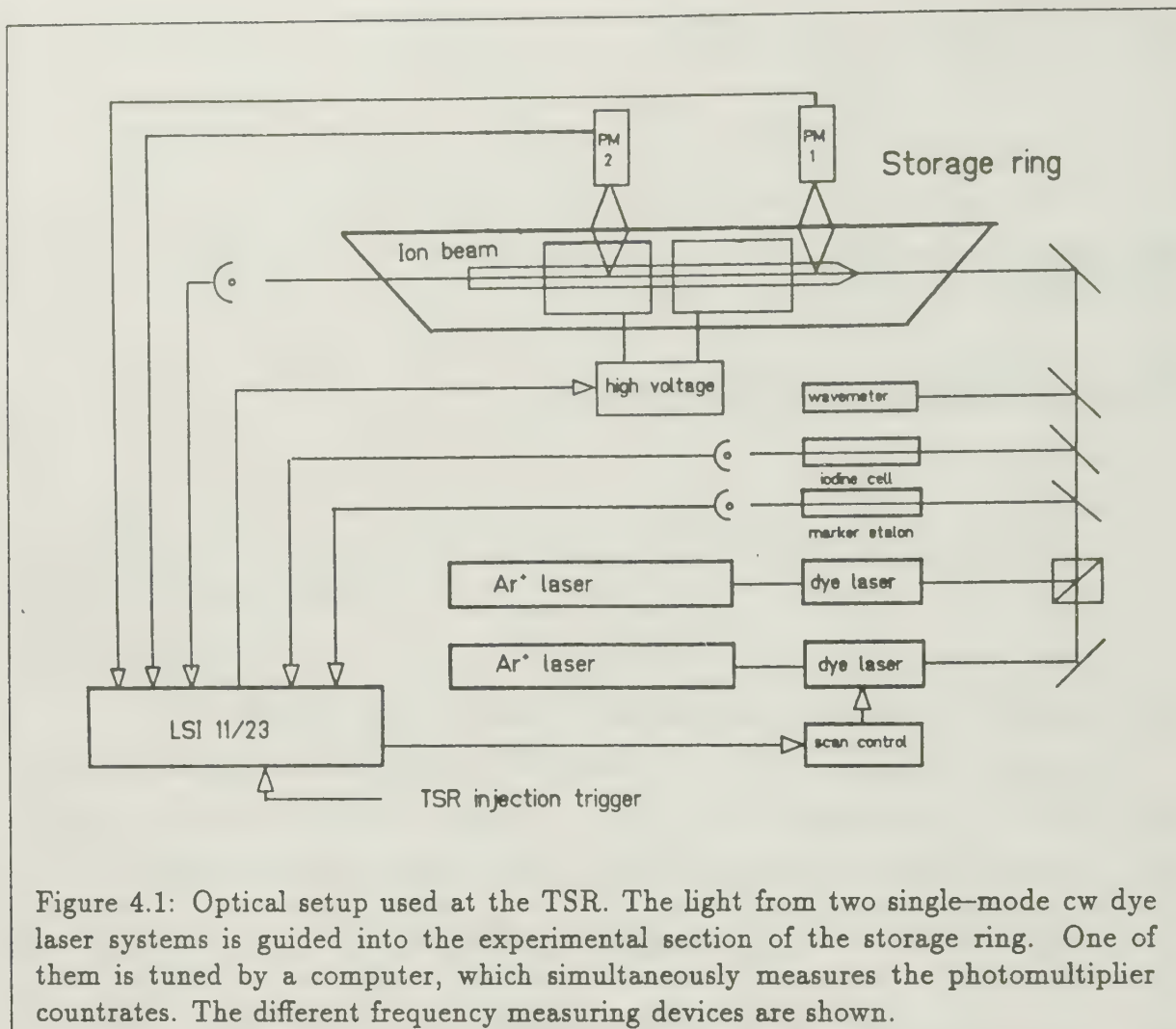


Figure 4.1: Optical setup used at the TSR. The light from two single-mode cw dye laser systems is guided into the experimental section of the storage ring. One of them is tuned by a computer, which simultaneously measures the photomultiplier countrates. The different frequency measuring devices are shown.

The linearity of a laser scan can be checked with a 300 MHz marker etalon.

Apart from a conventional beam transport to the TSR by high reflection mirrors, the two laser beams could be merged and coupled into one single mode optical fibre, delivered by Schott/Mainz, which provided collinear Gauß shaped laser beams in the experimental section of the TSR. The laser beams can be blocked by mechanical shutters. The beam diameter is expanded to about 1 cm whereas the estimated size of the ion beam is 2.2 cm in vertical, and 3.6 cm in horizontal direction. In case of using the blue shifted laser frequency, both laser beams are retroreflected behind the straight ring section. Excellent parallelism is achieved, when the reflected laser light again passes the optical fibre and, without an optical isolator, distorts the stable dye laser operation. An in situ supervision of the merging angle of both laser beams to an accuracy of $\leq 10^{-5} \text{ rad}$ is done by a diffraction technique described in [Gerh89].

Fluorescence perpendicular to the ion beam is detected by two photomultiplier tubes

and optical imaging systems. By PM1 (RCA 8850) a detection length of about 15 cm along the beam path is viewed with a mean solid angle of about 1.8%. The imaging is done by 150 mm diameter condensor lenses ($f=250$ mm), directly mounted onto a 150 mm glass window at a distance of 270 mm from the center of the beam tube. A light guide collects the fluorescence at the focus. The large Doppler shift allows to suppress laser stray light by an interference filter (550 ± 5 nm) and colour glasses (BG36, OG530). Most of the data shown here are taken with PM1. Only for voltage scanning fluorescence signals PM2 (EMI 9558 Q, mounted to a 35 mm quartz window at 210 mm from the beam tube center) is used because it is sensitive to a section within one electrode, where field inhomogenities are not very strong.

Doppler tuning is a convenient scanning method [Kauf76] in colinear fast beam spectroscopy. By applying up to 5 kV to two 0.67 m tube electrodes mounted close to each other in the middle of the 9.5 m experimental section of the TSR [Petr89], the ions inside the tubes are Doppler tuned by 1.5 MHz/V at 9.21 MeV and 1.3 MHz/V at 13.3 MeV, respectively.

Experiment control and data acquisition is done by an LSI 11/23 microcomputer and a Camac bus system. Measurements are mostly triggered by the TSR injection.

4.2 Ion Beam Properties

The detection of the Doppler broadened fluorescence line from ions stored in the TSR (fig. 4.2) immediately gives detailed information about ion beam and storage ring properties. However, there are some restrictions to the applications of these results, because our experiment is sensitive only to ions in the 3S_1 ($F = 5/2$) state and propagating within the laser beam. Besides this there are only those ions in the beam contributing to the optical signal, which are illuminated by the relatively small laser beam, compared to the about 4 times 2 cm ion beam. Experimental errors introduced by angle uncertainties are estimated below. From the measured fluorescence signal three quantities can be evaluated, which yield information about ion beam properties.

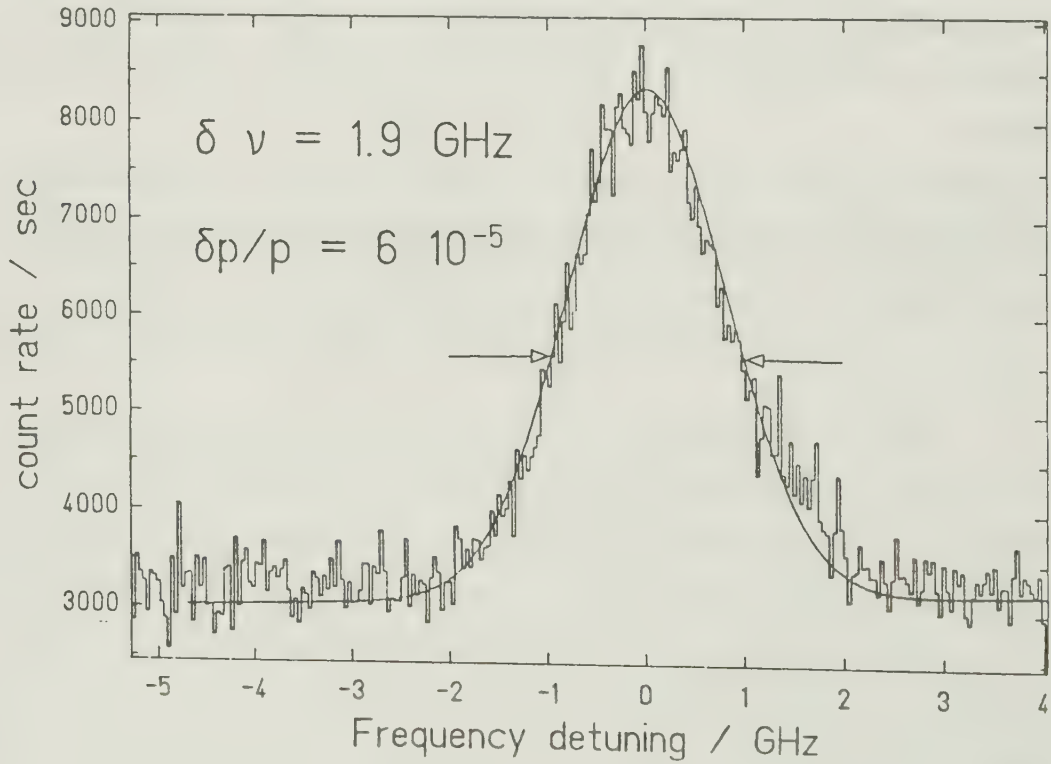


Figure 4.2: Fluorescence signal from the stored $^7\text{Li}^+$ ion beam. The laser is scanned from lower to higher frequencies around 17290.21 cm^{-1} to excite the $^3S_1(F = 5/2) \rightarrow ^3P_2(F = 7/2)$ transition. The total scanning time is 2.8 s.

• line center

- width of the Gauß profile
- signal amplitude

From the signal count rate we conclude that 10–30% of all stored ions are in the metastable 3S_1 state. The frequency ν_- , which corresponds to the line center, can be measured to an accuracy of $2 \cdot 10^{-7}$ given by the iodine molecular absorption lines we used as a reference [Gers78, Gers79].

$$\nu_- = 17290.207(3)cm^{-1} \quad \text{for antiparallel beam direction} \quad (4.2)$$

Since without optimization, in this measurement the angle Θ between laser and ion beam is below $5 \cdot 10^{-3}rad$, the ion's velocity can be calculated from the frequency Doppler shift via formula (3.7) to an accuracy of $1 \cdot 10^{-5}$. A simultaneous measure of the ion's revolution frequency with a pick up electrode ($f = 0.285730(7)$ MHz), determines the actual path length the ions travel around the storage ring $L = \beta \cdot c \cdot f^{-1}$.

$$\beta = 0.0527790(5) \quad (4.3)$$

$$L = 55.3767(14)m \quad (4.4)$$

With the known mass of $^7Li^+ mc^2 = 6.5349070(21) GeV$ [Waps85], we can also determine the kinetic energy $eU_{ac} = m_0c^2(\gamma - 1)$, which gives the accelerating voltage of the Tandem, taking into account the injection energy (190 keV) as well as the energy loss in the stripping process and by beam transport. This can be done with high accuracy and yields actually a high precision method for calibrating high voltages [Poul82].

$$U_{ac} = 9.1209(1)MeV \quad (4.5)$$

This energy measurement is by a factor of about 30 more accurate than the value given by the applied magnetic fields in the beam line. Finally the width of the fluorescence line is a direct measure of the longitudinal momentum spread $\delta p/p$ of the ion beam according to 5.15. From the spectrum shown in figure 4.2 we evaluate

$$\frac{\delta p}{p} = 6 \cdot 10^{-5} \quad (4.6)$$

This number is in agreement with Schottky noise measurements, but both are averaged values. Usually we average over several (typ. 10) subsequent ion injections, but a series of 13 single injection measurements yields information about the stability of the accelerator

and beam transport. The mean full width at half maximum (FWHM) of these fluorescence signals is only 1.26 GHz, equivalent to $\delta p/p = 3.6 \cdot 10^{-5}$, but the line center shifts statistically by $\pm 45\%$ the FWHM. This corresponds to $\delta p/p = \pm 1.6 \cdot 10^{-5}$ or an energy jitter of ± 0.43 keV. The width itself varies by $\pm 17\%$ the FWHM or $\delta p/p = \pm 6 \cdot 10^{-6}$. The numbers extracted from averaged measurements represent the statistical sum of these fluctuations.

The signal in figure 4.2 has not been corrected for the beam lifetime, since this is quite larger than the sweeping time of 2.8 s, during which the spectrum is taken. Doing so would pronounce the slight asymmetry of the fluorescence line even stronger. As the laser frequency is tuned towards higher values, that is in the cooling direction and decelerated ions may stay in resonance, this smoothing on the right side of the Doppler profile is already a first indication of momentum transfer by resonant photon scattering. By reversing the direction of the frequency sweep the shifting is avoided, and a clear example of this difference will be seen under experimental conditions optimized for laser cooling (see 6.4).

Tracing the time dependence of the three quantities listed above, allows to study the dynamical behaviour of the ion beam.

Recording the fluorescence count rate with fixed laser frequency on the $F = 5/2 \rightarrow F' = 7/2$ transition at low intensity, yields the storage lifetime of the metastable 3S_1 ions. Such a measurement is shown in figure 4.3, extending over more than one injection period of the TSR. However, the fluorescence does obviously not decay by a simple exponential curve, but may be approximated by a superposition of two exponential functions. The fast dropping of the count rate is interpreted as shifting of the originally resonant ions out of resonance due to photon momentum transfer. The particular decay times in the range of 150 to 500 ms for $20 \leq S \leq 60$ are in agreement with calculations including the mean light pressure acceleration derived in section 5.3. On a larger timescale ions are shifted back into resonance by velocity changing processes, resulting in a slow decay, which represents approximately the 3S_1 storage lifetime τ_m . A proper measurement of τ_m has to use small laser intensities to minimize the shifting, but the signal statistics gets worse rapidly. The best way is to perform several single measurements with different delay times from ion injection to switching on the laser beam.

The variation of the center and the width of the fluorescence signal with storage time is measured by series of fast adjacent laser sweeps, scanning the line profile four times

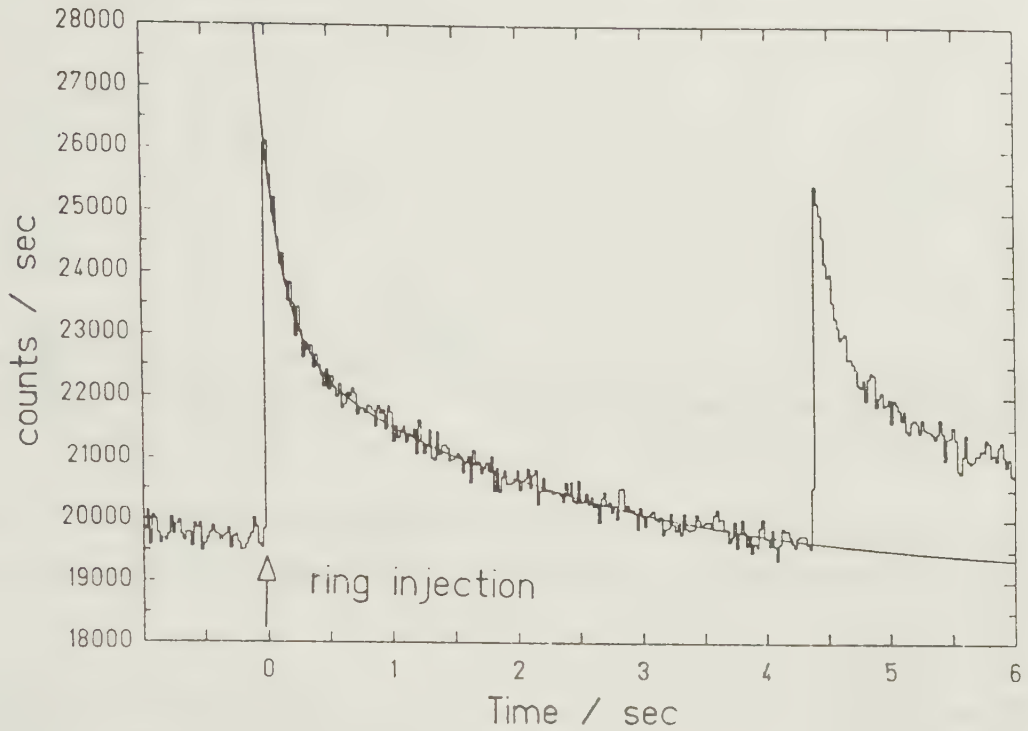


Figure 4.3: Fluorescence signal at fixed laser frequency. The decay may be approximated by two exponential functions representing the light pressure shifting ($\tau = 200$ ms) and the metastable beam lifetime ($\tau = 5$ s).

within one storage lifetime. The time diagram is shown in figure 4.4. The laser beam is blocked during the reset of the frequency. From the areas of the fourfold measured fluorescence line the beam lifetime may be obtained. The results fit reasonably well with the time constants of the slow decay curves described above. The evaluated line centers and widths are plotted in figure 4.5 and show clearly, that the shift of the line center as well as the increase of the linewidth is dominated by residual gas scattering. Three different situations are realized. The data points indicated by 'residual gas' are taken by blocking the laser beam for the first frequency sweeps, that is after different delay times from ion injection. The other two series differ by the direction of the laser frequency sweep. The line center is shifted slightly more when scanning in the cooling direction (red to blue) due to the shifting of ions by multiple photon momentum transfer. However, the main influence arises from residual gas collisions.

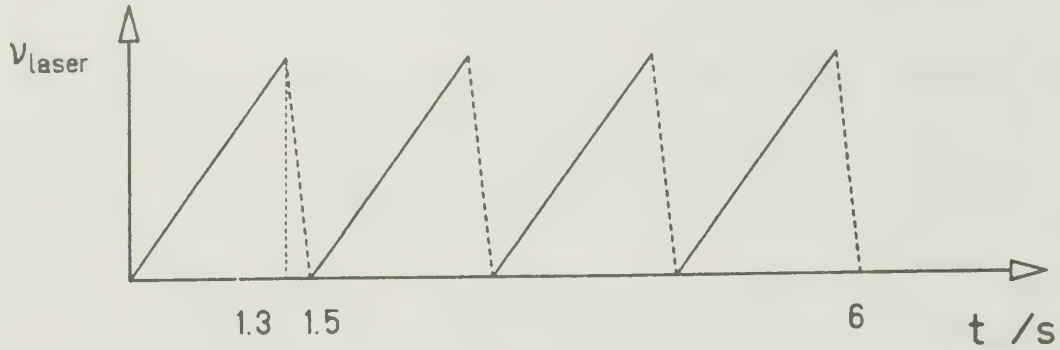


Figure 4.4: Time diagram of fourfold laser scan. The fluorescence signal is measured during the long frequency slopes and the laser beam is blocked during the frequency reset.

- The line center is shifted by a continuous slowing down of the ion beam. This shift of 150 MHz/s at a residual gas pressure of $7 \cdot 10^{-10}$ mbar corresponds to an energy loss of 100 eV/s or a velocity change of 85 m/s^2 .
- Besides this shift, the multiple small angle scattering also leads to a straggling or line broadening in the order of 200 MHz/s equivalent to a growth of the relative momentum spread of $\frac{\delta p}{p} \approx 7 \cdot 10^{-6}/s$.

These data show, that the narrow initial momentum spread of the ion beam does not increase strongly. Line shift and width are in qualitative agreement with small angle multiple scattering within the theory of Lindhardt et.al. [Lind63, Sigm74, Side75]. The energy loss of the beam can be deduced from the tabulated data in [Nort70]. For a residual gas composition of 50% H_2 and 50% N_2 at a total pressure of $3 \cdot 10^{-8}$ Pa, a loss of 104 eV/s or an equivalent frequency shift of 156 MHz/s is predicted.

The energy straggling due to multiple Coulomb scattering is calculated for internal targets in storage rings by [Hint89]. Taking the results over for residual gas scattering, the rms energy spread ΔE_{rms}^2 after a target of areal density ρx is given as

$$\Delta E_{rms}^2 = 0.1535 \frac{\text{MeV cm}^2}{g} \frac{Z_1^2 Z_2}{\beta^2 A_2} \cdot \rho x \cdot \Delta E_{maz} \left(1 - \frac{\beta^2}{2} \right) \quad (4.7)$$

Z_i and A_i are atomic and mass numbers of the colliding partners and ΔE_{maz} denotes the maximum energy loss in a head-on collision with a target electron. This number is also calculated and we get 2.8 keV for our experimental parameters. Thus during 1 s and $1.5 \cdot 10^9$ cm of traversed residual gas at the conditions given above a mean energy spread

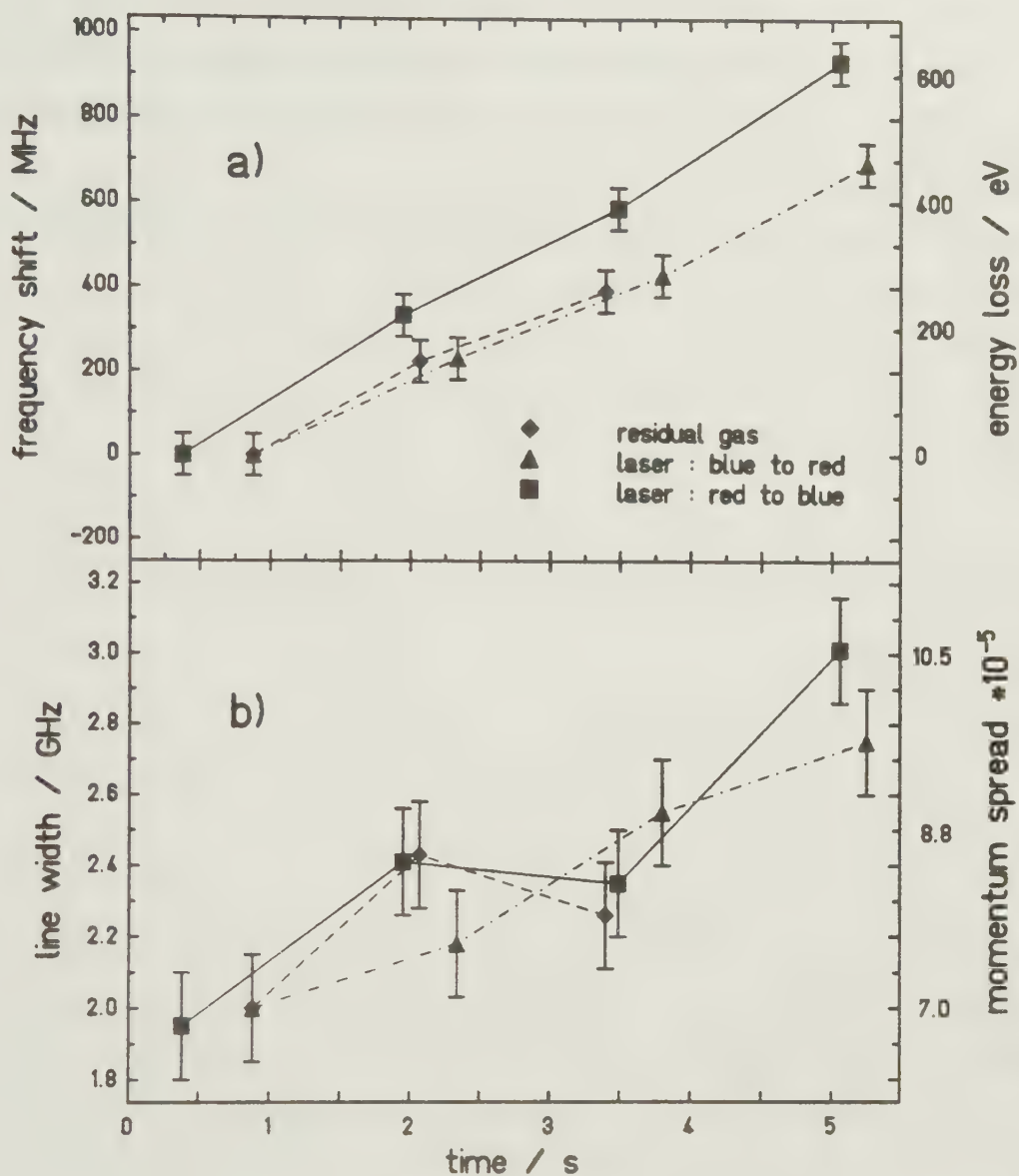


Figure 4.5: Shift of the line center (a) and line widths (b) as a function of storage time. In comparison to data taken at different directions of laser frequency scanning, a delay measurement is shown (marked 'residual gas'), where the laser beam is blocked for the first sweeps.

of 410 eV is calculated, which is by a factor of two more than measured. However it is in reasonable agreement regarding the uncertainties in the vacuum parameters.

Thus we conclude that the intrabeam scattering among the ${}^7\text{Li}^+$ ions has a neglectable effect on the line broadening at our beam intensities, beam size and momentum spread. Otherwise the large transverse kinetic energy of the beam would be transferred

to the longitudinal motion by Coulomb collisions between ${}^7\text{Li}^+$ ions [Bjor83, Cont85] and a dramatic increase of the fluorescence line width would be observed. This is also consistent with theoretical intrabeam scattering calculations, done by [Wolf88] for a very similar ${}^9\text{Be}^+$ beam. Adopting these to the ${}^7\text{Li}^+$ case, yields longitudinal energy spreads in the order of few meV/s.

Residual gas scattering, however, represents a considerable heating mechanism in the TSR.

4.3 Particle Dynamics in the Ring

Even more detailed information about ion beam kinematics in a storage ring may be gained by detecting the behaviour of one specific velocity class. By optical pumping with a fixed frequency laser, ions with well defined longitudinal velocity v_z are marked. We chose the V - system, inserted in figure 4.6, connecting the 3S_1 ($F = 5/2$) with the 3P_2 ($F' = 7/2$) and ($F' = 5/2$) levels, separated by 11.76 GHz¹. Two copropagating laser beams are directed towards the oncoming ion beam. While the $5/2 \rightarrow 7/2$ transition is scanned by the first laser, the $5/2 \rightarrow 5/2$ transition is excited by a fixed frequency second laser. Ions in resonance with this second laser are optically pumped out of the active $^3S_1(F = 5/2)$ ground state after 1.4 excitations on average.

As a result the observed fluorescence line shows a Lamb dip (fig. 4.6 a)) and we analyze it's

- width,
- amplitude and
- relaxation time

as a function of the laser intensity I_{pump} used for excitation of the $5/2 \rightarrow 5/2$ pumping transition. Figure 4.7 shows the power dependence of the dip width produced by a laser beam with 7.4 mm diameter. The width $\delta\nu$ is plotted quadratically against I_{pump} , because of the expected square root dependence $\delta\nu = \delta\nu_{nat} \sqrt{1 + I_{pump}/I_{sat}}$ of this saturation effect.

On behalf of a qualitative understanding of these Lamb dips, the experiment is simulated theoretically . The three level system (1)=($F=5/2$), (2)=($F'=5/2$) and (3)=($F=3/2$) is described by the rate equations

$$\dot{n}_1 = -n_1 w + n_2 (A_1 + w') \quad (4.8)$$

$$\dot{n}_2 = -n_2 (A + w') + n_1 w \quad (4.9)$$

$$\dot{n}_3 = n_2 A_2 \quad , \quad (4.10)$$

where n_i is the population of level (i), A is the spontaneous decay rate of the excited level $F'=5/2$, splitting in A_1 for the decay back to $F=5/2$ and A_2 to $F=3/2$. The excitation rate w of $F = 5/2 \rightarrow F' = 5/2$ is equal to the reverse induced emission rate w' . Solving

¹For application of an optical Λ -system see appendix C

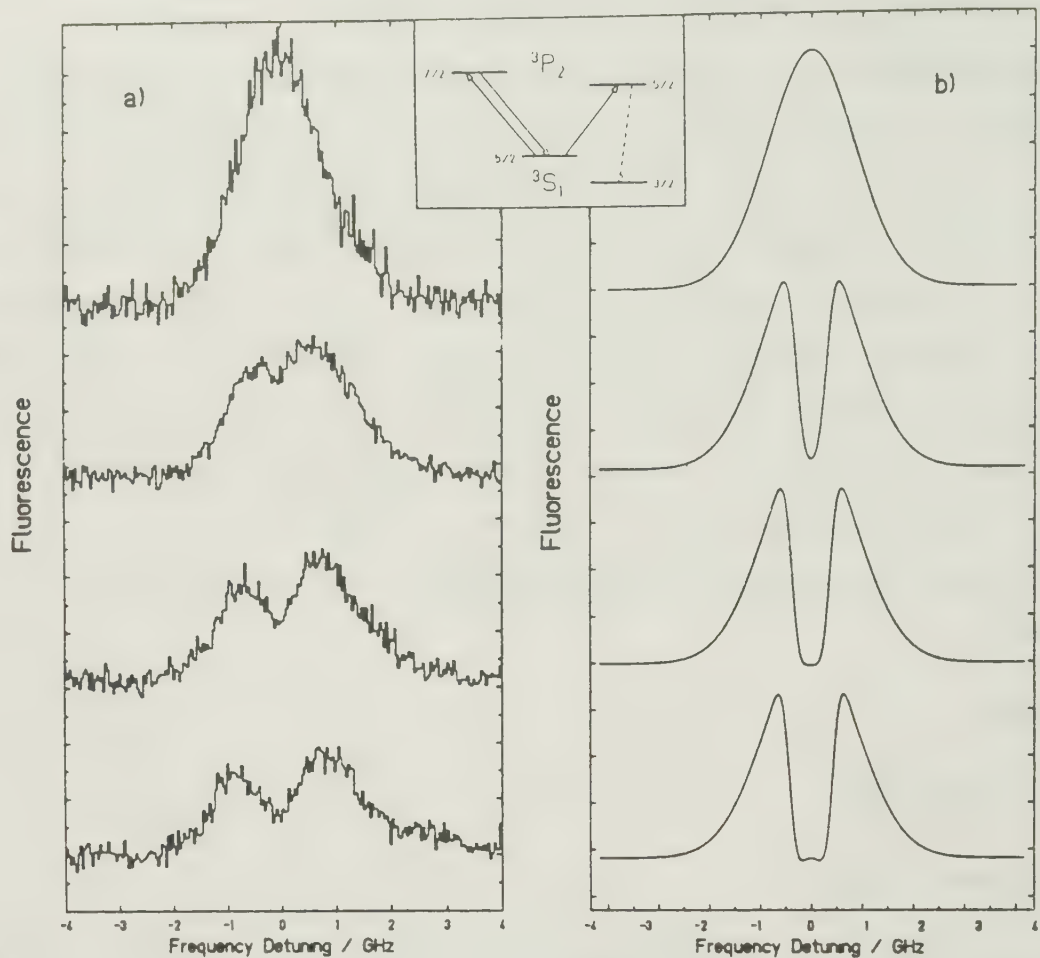


Figure 4.6: Lamb dip at different pumping laser intensities. The original fluorescence line without optical pumping is shown for comparison above in (a). The lower spectra are taken after optical pumping on the $5/2 \rightarrow 5/2$ transition at laser intensities of 1, 2.4 and 4.8 mW/cm^2 , respectively. Rate equation calculations of the line shape are plotted in (b) for the same experimental settings. These, however, do not take the geometrical overlap of the beams into consideration.

these equations under the assumption of small laser intensity in the pumping transition ($w \ll A$) leads to

$$n_2(t) = e^{-\frac{A_2}{A}wt} + \frac{wA_1}{A^2} e^{-At} \quad (4.11)$$

Because induced emission is neglected, the observed fluorescence (from both decay branches) is proportional to $n_2(t)$, where t is the effective interaction time of the ions with the pumping laser radiation. The excitation rate w has to be calculated by convolution

of the lorentzian frequency dependence of the optical transition $L(\nu)$ and the gaussian broadening effect $G(\nu_B)$ due to the betatron oscillations (see section 5.3). It is assumed, that the laser bandwidth is smaller than the natural width $\delta\nu_{nat}$ of the transition.

$$w = S A \int_{-\infty}^{+\infty} L(\nu) \cdot G(\nu - \nu_B) d\nu \quad (4.12)$$

This integral has been solved numerically and the results of the Lamb dip calculations are shown in figure 4.6b) for similar laser intensities as used in the measured spectra 4.6a). The two parameters interaction time t and betatron oscillation width $\delta\nu_B$ are fitted to match the absolute values and the power slope of the dip widths (see fig. 4.7).

$$t = 200\mu s \quad (4.13)$$

$$\delta\nu_B = 400MHz \quad (4.14)$$

The effective interaction time of only $200\mu s$ is by a factor of $2 \cdot 10^{-4}$ smaller than the actual time of 1 s, for which the pumping laser is on before the Lamb dip is probed by the scanning dye laser. This reduction of the ion interaction time with the laser light is due to different effects. The duty cycle due to the ion circulation around the ring, the geometrical, and the frequency overlap are considered quantitatively in section 5.3, giving a total reduction in the order of 10^{-4} . The factor here seems to fit very well with this value, but we will have to apply small corrections below.

The extrapolated zero power width, which is approximately the induced line width due to the betatron oscillations (compare sec. 3.1), is unexpectedly large for a saturation spectroscopic experiment, where Lamb dips are usually in the order of the natural line width [Demt88]. In contrast to the calculated spectra the measured Lamb dips do not drop down to the noise level (fig. 4.6), which may also be seen from figure 4.7b, where the relative depth is plotted. Different reasons may be quoted to explain this effect.

- Ions with betatron oscillations in the order of the laser beam diameter are pumped much more effectively than those with large oscillation amplitudes (see sec. 5.3). Thus the latter may contribute to the background fluorescence in the region of the Lamb dip. This effect of spatial overlap with the laser beam has not been included in the simulation, because due to the large uncertainties in the actual size, position and angular orientation of the ion beam a much more detailed analysis concerning the experimental geometry would be necessary. A special computer code for single

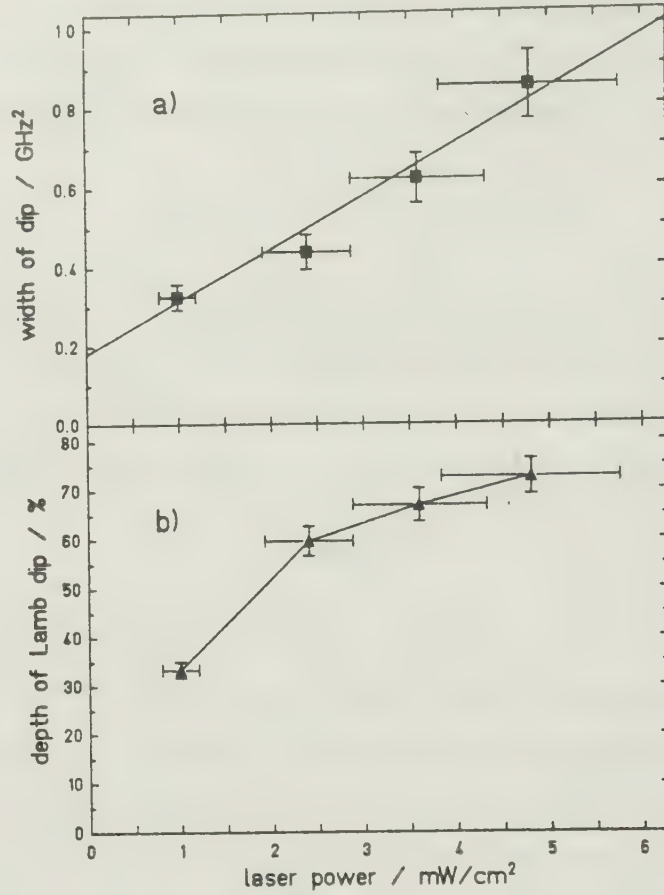


Figure 4.7: The power dependence of the width (a) and the depth (b) of the Lamb dips is shown. Results from rate equation calculations are included as linear dependence in (a).

ion tracing in the TSR is in preparation [Klei89].

- Population mixing (see 3.3) and velocity changing in the storage ring will also contribute to this background but are excluded from the simulation as well.

The velocity mixing time scale in the TSR is of fundamental interest for precision experiments as well as for laser cooling. The direct measurement of the lifetime τ_v of velocity information within the ion beam is done by recording the fluorescence signal in time with both lasers fixed to the $5/2 \rightarrow 7/2$ and $5/2 \rightarrow 5/2$ resonance respectively and by turning the optically pumping laser off after some time. The quickly decreasing count rate then recovers and the ion velocities are redistributed statistically (fig. 4.8). The measured relaxation times, which correspond to about $3 \cdot 10^4$ revolutions of an ion around the storage ring, are plotted in figure 4.9. This effect may be partly due to population mixing of the 3S_1 F-substates, where timescales below 1 s for optical pumping are estimated (see sec.

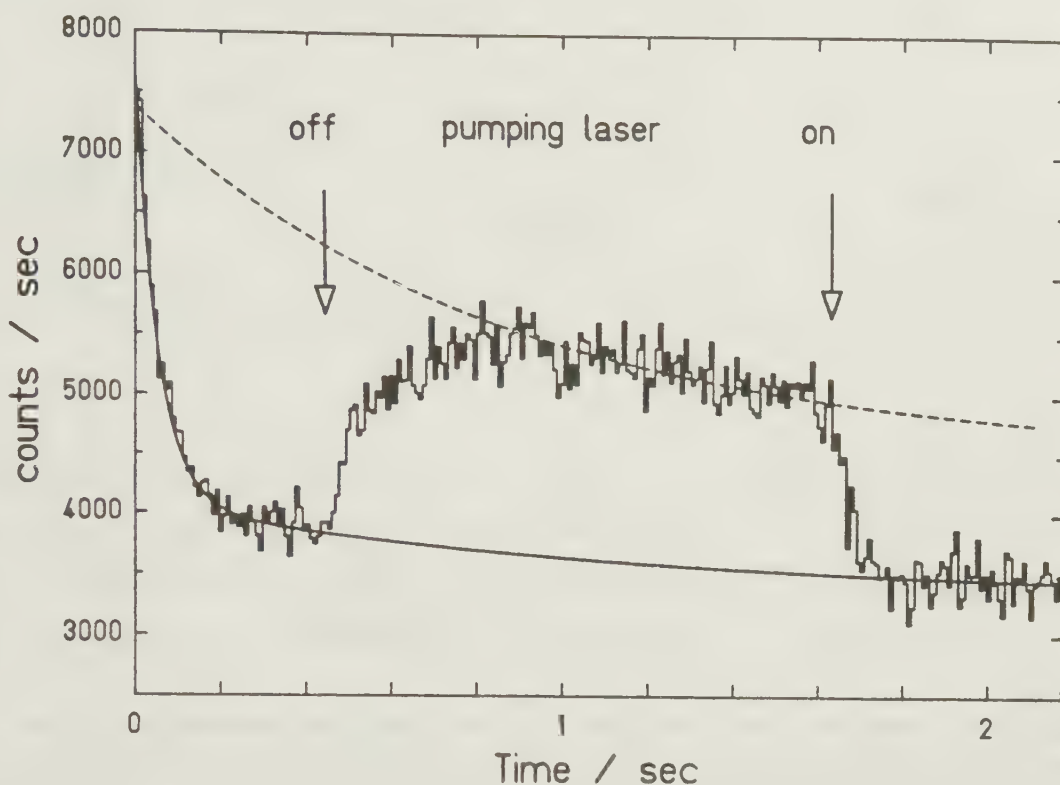


Figure 4.8: Measured fluorescence with both lasers fixed on the pumping and probing transition respectively. Blocking the weak pumping laser beam lets the fluorescence recover within typically 100 ms.

3.3). On the other hand also velocity changing processes in the storage ring contribute to the recovering of the fluorescence signal in figure 4.8. The fact, that — without any further stabilization — deviations from the gaussian momentum distribution of the stored ion beam are washed out after typically $\tau_{dip}=100$ ms, implies a steady state between optical pumping and a momentum relaxation for the Lamb dip measurements. Thus the effective interaction time t has to be compared with these 100 ms. The corresponding reduction factor is only $2 \cdot 10^{-3}$. This may be explained by the privileged interaction of the laser light with ions having small betatron oscillation amplitudes.

The long lifetime of a Bennett hole, as well as the narrow longitudinal momentum spread itself, prove that for these experimental conditions interactions between ions in the beam like intrabeam scattering and any kind of inelastic collisions (excitation transfer, charge exchange) may be neglected. Collision rates can be estimated to be lower than 10 Hz, and thus the $^3S_1(F = 5/2)$ ions may be regarded as isolated on this timescale,

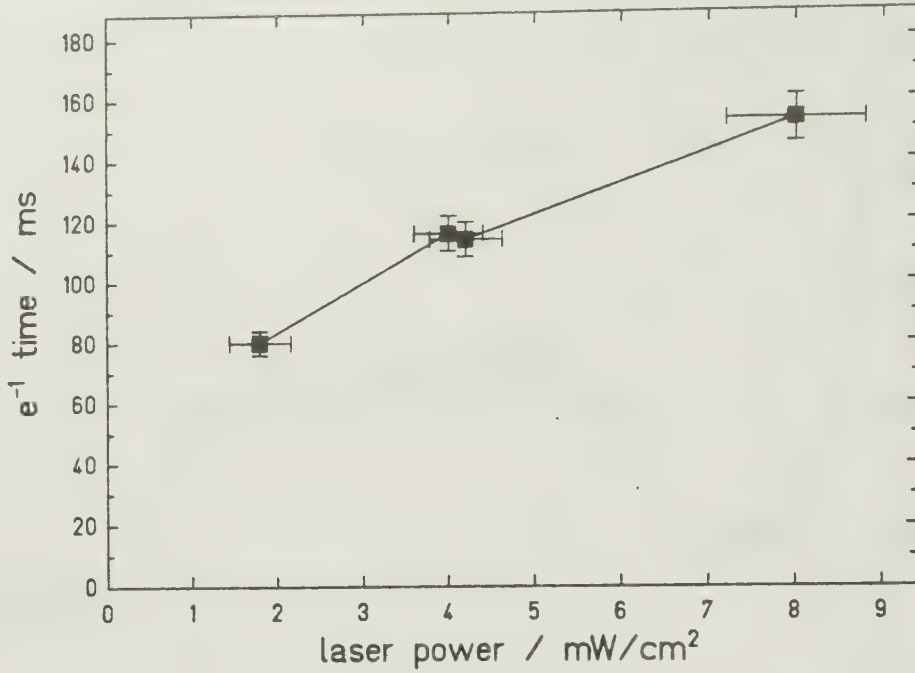


Figure 4.9: Measured time constants for the fluorescence recovering. These time constants are identified with the lifetime of velocity information in the TSR.

that means no effective sympathetic cooling is present.

The results from ion beam diagnostics as described up to now, show the feasibility of applying laser cooling to a ${}^7\text{Li}^+$ ion beam in the TSR. The precondition is fulfilled, that the lifetime of velocity classes in the ion beam is long compared to the natural lifetime prolonged by the interaction time reduction factor.

$$\tau_{\text{dip}} = 100\text{ms} \gg \frac{\tau_{\text{nat}}}{2 \cdot 10^{-4}} = 0.2\text{ms} \quad (4.15)$$

In the next chapters a brief report of the theory of laser cooling will be given and the experimental results obtained at the TSR are presented.

Chapter 5

Ion Beam Cooling

A universal quantity describing the internal motion of a number of particles in the rest frame of their mean velocity is the ensemble temperature. For a thermal velocity distribution with $|\vec{v}|$ following the Boltzmann statistics the distribution of the one-dimensional projection v_z is Gaussian. In this case the temperature T can be defined via

$$\frac{1}{2}k_B T = \frac{1}{2}M(\delta v_z)^2 \quad , \quad (5.1)$$

where δv_z is the half width at half maximum of the velocity distribution, M is the particle mass and k_B denotes the Boltzmann constant.

In the case of the ${}^7\text{Li}^+$ beam stored in the TSR the longitudinal velocities are Gaussian distributed due to the statistical production process of the ions in the accelerator. The velocity of the beam is actually given in terms of the relative momentum spread $\frac{\delta p}{p}$ or, when measured by laser spectroscopy, by the optical frequency width $\frac{\delta \nu}{\nu_-}$, and thus the beam temperature may be calculated according to

$$T = \frac{Mc^2}{4k_B} \beta^2 \left(\frac{\delta p}{p} \right)_{FWHM}^2 = \frac{Mc^2}{4k_B} \left(\frac{\delta \nu}{\nu_-} \right)_{FWHM}^2 \quad , \quad (5.2)$$

where $\delta \nu$ is the full width of the fluorescence peak and ν_- the red shifted resonance frequency. The measured momentum spread of $6 \cdot 10^{-5}$ (see sec. 4.2) corresponds to a longitudinal temperature $T_{||}$ and if the transverse motion due to the betatron oscillations is also expressed as a temperature $T_{\perp}$, we realize as the initial conditions at the TSR

$$T_{||} = 260 K \quad (5.3)$$

$$T_{\perp} \approx 10^5 K \quad (5.4)$$

5.1 Stochastic and Electron Cooling

The first cooling technique developed at elementary particle storage rings at CERN is the stochastic cooling [Meer85]. The electronic signal induced by a stored particle at a differential pick-up electrode in the ring is amplified and fed to a kicker electrode in a different section of the ring. The signal should arrive at the kicker at the same time as the particle and this feedback circuit will damp its transverse motion. Using longitudinal pick-up electrodes, the longitudinal motion can also be cooled. Although for many particle beams this technique introduces some heating, the principle of operation remains and the electronic parameters can be chosen to get a net cooling effect. Stochastic cooling works up to highest beam intensities and is also implemented at the heavy ion storage ring ESR.

At the TSR an electron cooling device is implemented for improving the phase space density of stored ion beams [Stec90]. A cold electron beam of 5 cm diameter and a maximum current of 1 A is merged with the stored ion beam on a length of 1.5 m. The velocities of electrons v_e and ions v_{ion} have to be matched in order to get an effective heat exchange between the two media. This condition may be expressed in terms of energies

$$v_e = v_{ion} \Leftrightarrow E_{e,kin} = \frac{m_e c^2}{M_{ion} c^2} E_{ion,kin} \quad , \quad (5.5)$$

with m_e and M_{ion} the rest masses of electron and ion. For 9.21 MeV ${}^7\text{Li}^+$ ions, the electrons have to be accelerated to only 721 eV, which is much below the optimum operating conditions of the electron cooler. Theoretically the minimum ion temperature to reach is defined by the electron temperature via the mass ratio

$$T_{ion} = \frac{m_e}{M_{ion}} \cdot T_e \quad , \quad (5.6)$$

but different heating mechanisms in the storage ring lead to experimental values of about one Kelvin.

A crucial question to the feasibility of electron cooling in our case, however, arises due to the slow collisions between ${}^7\text{Li}^+$ ions and electrons, which may quench the 3S_1 state. From electron bombardment measurements the cross section of the $1s^2 \, {}^1S_0 \rightarrow 1s2s \, {}^3S_1$ reaction of $\sigma \leq 2 \cdot 10^{-18} \text{ cm}^2$ is known [Roge78]. The detailed balance argument shows that the inverse reaction $e^- + {}^3S_1 \rightarrow e^- + {}^1S_0$ has a yield of maximum $4 \cdot 10^{-2} \text{ s}^{-1}$ for a 0.08 A/cm^2 electron beam comoving to the 9.12 MeV ${}^7\text{Li}^+$ beam. Thus the 3S_1 state is not quenched by the electron cooling beam, which has been verified experimentally within

a wide range of electron energies or relative velocities [Schr88a, Schr88b] (see appendix B).

The cooling time required for electron cooling of ${}^7\text{Li}^+$ may be scaled from the measured value $\tau_{\text{ecool}}=2$ s for a proton beam [Habs89]. According to $\tau_{\text{ecool}} \propto \frac{A}{Z^2}$ (A-mass number, Z-charge state) [Habs89] we expect 14 s for electron cooling of a ${}^7\text{Li}^+$ beam, which, however, is longer than the 3S_1 storage lifetime.

Both these established cooling methods at storage rings are limited in their performance to temperatures in the Kelvin region. The new laser cooling technique, which is applied here for the first time to an ion beam in a storage ring, has yielded drastically lower temperatures in the μK regime in atomic beam cooling experiments. The main features of the application of laser cooling to fast stored ions will be pointed out in the next sections.

5.2 Laser Cooling of Free Atoms and Ions

The theory of laser cooling as developed by different authors [Cook79, Leto81, Fene77, Sten86] may be applied to free atoms as well as ions. However, first experimental investigations concentrated on thermal atomic beams cooled longitudinally by overlapping with a colinear laser beam [Andr81, Phil82].

The effect of laser cooling utilizes the momentum transfer connected with the absorption of a photon by an atom. In order to produce a macroscopic change in the atom's velocity, many of these momenta of size $\hbar\vec{k}$ need to be added up in one defined direction. Therefore it is necessary, that the atom to be cooled shows a closed two level system of quantum states with the lower one being the atomic ground state or a long lived metastable state. Considering a two level atom with transition energy $\hbar\omega_0$ interacting with an electromagnetic wave near resonance, spontaneous transitions occur as well as stimulated ones. Besides fully quantum mechanical treatments of the connection of these internal to the translational degrees of freedom [Fene77], most derivations of the mechanical effects of light on free atoms use the semi-classical approach [Leto81]. Here the translational atomic motion is considered classically, which is justified since the atom's de Broglie wavelength λ_{DB} is considerably less than the light field wavelength λ .

$$\begin{aligned} \lambda_{DB} = \frac{h}{p} &\ll \lambda = \frac{2\pi}{k} \\ \Leftrightarrow p &\gg \hbar k \end{aligned} \quad (5.7)$$

where p is the atomic momentum and $k = |\vec{k}|$ the wavevector of the light field. In terms of energy this condition is equivalent to a kinetic energy E_{kin} of the atom being much larger than the recoil energy R due to one photon absorption.

$$E_{kin} = \frac{p^2}{2M} \gg R = \frac{\hbar^2 k^2}{2M} \quad (5.8)$$

Here M is the atomic mass. Looking at the time scales of both processes, we are in the regime of adiabatically slow variation of the atomic velocity in comparison with the time for excitation or spontaneous decay of the atomic eigenstate. For a natural line width of the transition of Γ the condition may be written as

$$\hbar\Gamma \gg R \quad (5.9)$$

This additional argument allows us to describe the atomic motion as influenced by a

classical force.

Detailed calculations show, that the total light pressure force $\vec{F}_{lp}$ can be written as a sum of two components

$$\vec{F}_{lp} = \vec{F}_{sp} + \vec{F}_d \quad . \quad (5.10)$$

The spontaneous force $\vec{F}_{sp}$ is directed along the axis of the light wave and results from absorption and spontaneous reemission of photons at a rate given by the saturation parameter $S = I/I_S$, where I and I_S denote the actual and the saturation intensity of the driving laser field, respectively. As the mean momentum of the emitted photons is equal to zero ($\langle \Delta v_{sp} \rangle = 0$), the net effect to first order is a unidirectional velocity change (see fig. 5.1) expressed as a force

$$\vec{F}_{sp} = \frac{\hbar \vec{k} \Gamma}{2} \frac{S}{1 + S + 4(\Delta - \vec{k} \cdot \vec{v})^2 / \Gamma^2} \quad , \quad (5.11)$$

where $\Delta = \omega - \omega_o$ is the detuning from exact resonance. F_{sp} is a dissipative force, which saturates with increasing laser power and has a lorentzian frequency dependence with full width at half maximum of $\Gamma \sqrt{1 + S}$. The maximum value of F_{sp} at high saturation is $\hbar k \Gamma / 2$. The momentum diffusion due to a non vanishing variance $\langle (\Delta v_{sp})^2 \rangle > 0$ associated with the spontaneous force will be discussed below.

The second term in equation 5.10 results only from induced absorption and emission processes. If we deal with a physical laser beam with radially decreasing intensity rather than a plane wave, the gradient or dipole force $\vec{F}_d$ is a consequence of the redistribution of momenta between the partial waves with $\vec{k}$ - vectors varying slightly in orientation. $\vec{F}_d$ is directed radially along the field gradient of the light beam.

$$\vec{F}_d = \frac{1}{2} \hbar (\Delta - \vec{k} \cdot \vec{v}) \frac{\nabla S(r)}{1 + S(r) + 4(\Delta - \vec{k} \cdot \vec{v})^2 / \Gamma^2} \quad (5.12)$$

Here $S(r)$ is the spatially varying saturation parameter, which is correlated to the Rabi flopping frequency in the two level system $\Omega = dE/\hbar$ (d atomic transition dipol moment, E electric field strength) via

$$S(r) = \frac{2\Omega(r)^2}{\Gamma^2} \quad . \quad (5.13)$$

The sign of $\vec{F}_d$, that is the direction of the force, depends on the detuning of the driving laser frequency from resonance. For $(\Delta - \vec{k} \cdot \vec{v}) < 0$ the atom is drawn into the light beam and vice versa. The dipole force is a conservative force, as it does not include any dissipative properties.

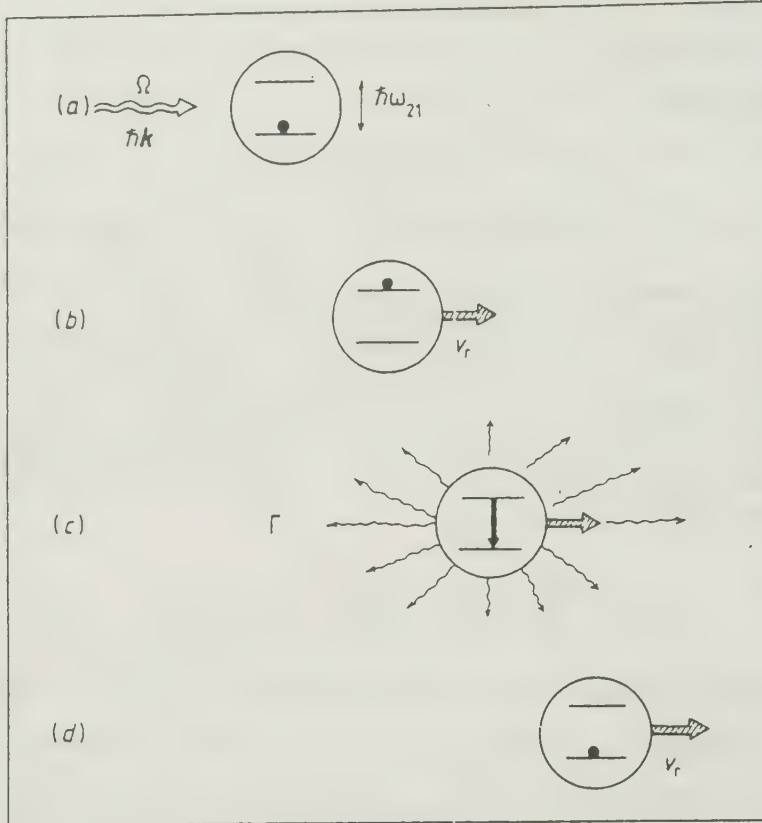
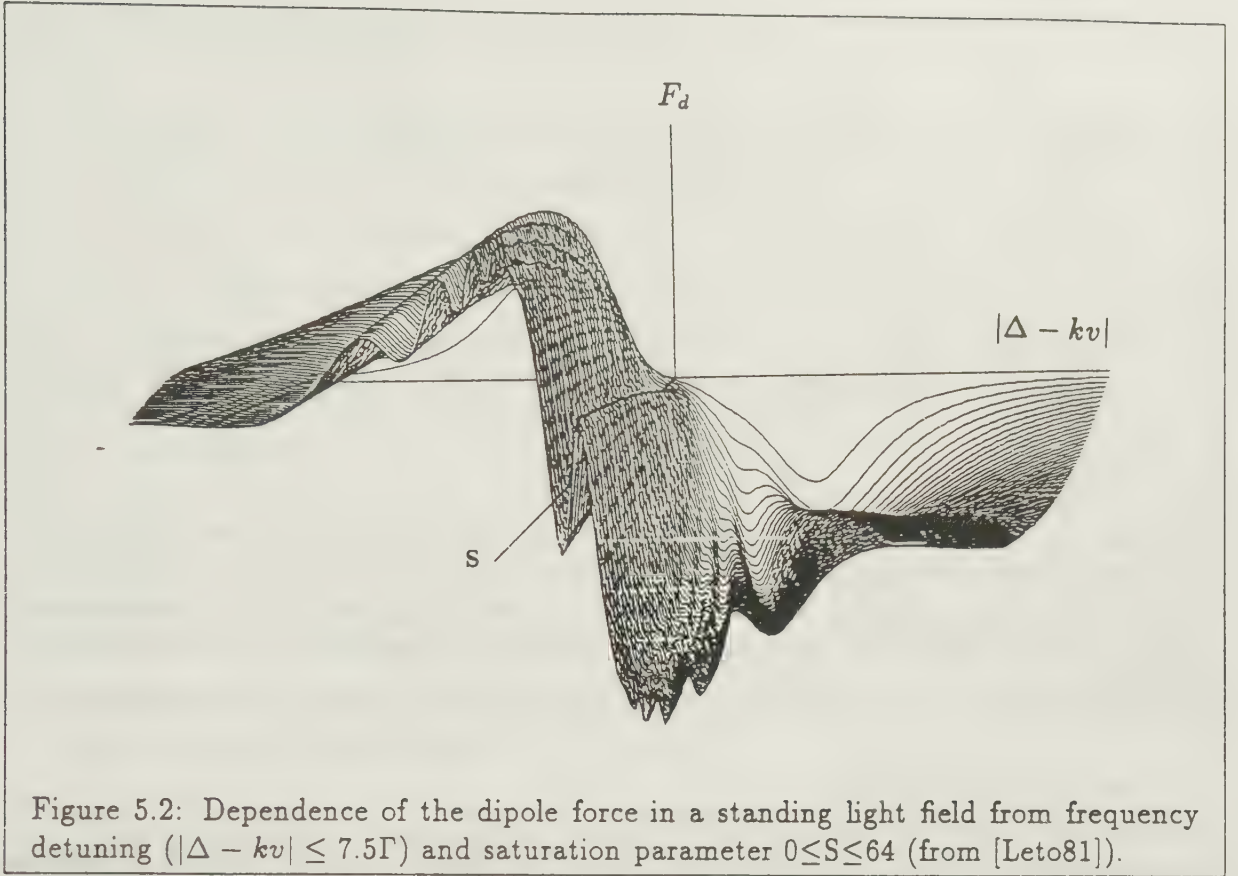


Figure 5.1: Elementary process leading to the spontaneous light pressure force. An ion with two internal energy levels is acted upon by laser light (a). Absorbing a photon from the light field the ion is excited (b) and takes the momentum $\hbar k$ over, resulting in an increase of the ion's velocity by v_r . The upper level decays spontaneously (c) at a rate Γ , but the angular distribution of the corresponding momenta average to zero in the laser beam direction. Finally the ion has returned to its ground state and has changed its longitudinal velocity by v_r (d) (from [Sten88]).

In an experimental situation, where the atom is moving in a standing light wave, realised e.g. by two counterpropagating laser beams, $\vec{F}_d$ also has a component along the light propagation axis. This is due to the strong variation of light intensity from node to antinode of the standing wave and has been described very successfully by the 'dressed atom' approach [Dali85]. The atomic motion along the axis is damped for detunings $(\Delta - \vec{k} \cdot \vec{v}) > 0$ and accelerated for $(\Delta - \vec{k} \cdot \vec{v}) < 0$. The qualitative dependence of F_d from the detuning is shown in fig 5.2. The dispersive behaviour is disturbed for high saturation parameters. Especially the inversion of the sign of F_d near zero detuning will lead to two stable points for different velocity classes, separated by only few MHz. Describing the experiments of this work I will restrict the discussion mainly to the spontaneous light pressure force, because the complex situation in a storage ring renders many difficulties



to the analysis of phase-dependent ion interaction with the laser fields.

Similar to thermal atomic beam experiments, the ion beam stored in the TSR shows a Gaussian distributed velocity profile. Due to the Doppler effect this gives an inhomogeneous broadening of the ionic transition of 2 GHz. As the action of F_{op} is limited to a narrow region in frequency space (the saturation broadened natural linewidth), tuning the cooling laser frequency to some position within the Doppler profile of the ion beam will result just in a distortion of the velocity distribution in a narrow frequency range. In order to collect all ions, that is all velocity classes in the beam, the velocity profile has to be scanned in a way, that the shifted ions are accumulated in a narrow peak in front of the laser frequency position. Figure 5.3 illustrates the technique of 'Doppler cooling'. Naturally the integral of the cooled and uncooled distribution should be equal, unless there are any loss processes present. Basically different schemes may be used to perform the sweeping.

1. The laser frequency may be tuned from lower to higher frequencies for counterpropagating laser and ion beams. Due to the decreasing Doppler shift, decelerated ions will stay in resonance with the shifting laser frequency (for application to atomic

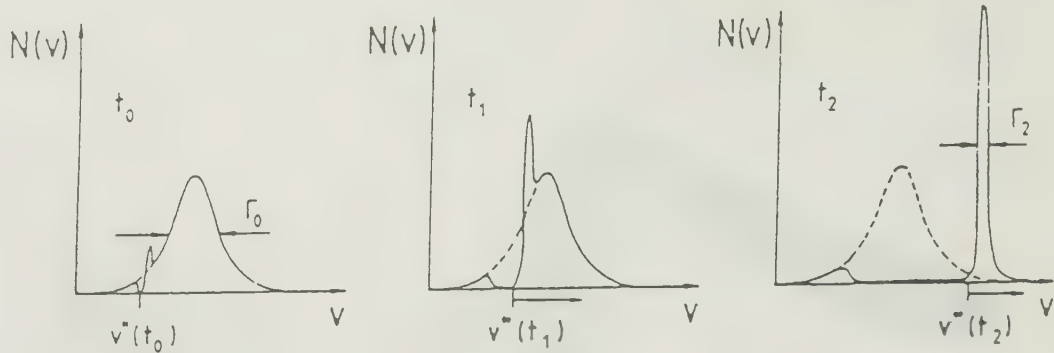


Figure 5.3: Doppler cooling technique. At time t_0 the velocity class $v^*(t_0)$ is in resonance and by sweeping the cooling laser frequency across the velocity distribution of the ion beam at time t_2 the ions are shifted and compressed by the spontaneous light pressure force. The width of the distribution is decreased from Γ_0 to Γ_2 (from [Petr89]).

beams see [Ertm85]).

2. At fixed laser frequency the atomic transition energy may be changed by Zeeman tuning in a varying magnetic field [Phil85].
3. For an ion beam with mean velocity $\langle v \rangle \gg 0$ 'Doppler tuning' may be applied via a high voltage electrode, where the ion's velocity and consequently the Doppler-shifted resonance frequency is changed [Java85].
4. In the TSR storage ring smooth de- and acceleration, and thus Doppler tuning, will be possible with a new induction accelerator device [Elle90], which, however, has not been operational for the experiments described in this work.
5. A similar effect of small range energy tuning can be achieved by changing the energy of the electron cooling beam. Within the effective range of the electron cooling force the ion beam will assimilate its velocity to that of the electrons.

Both possibilities 1 and 3 have been used in these experiments.

Thermal atomic beam cooling is performed within times of typically 1 ms on a length of 1 m. The essential changes for the application of laser cooling to relativistic ions in a storage ring will be pointed out next.

5.3 Laser Cooling of Stored Ions

Probably the most important difference of laser cooling of stored ions in comparison to the situation in atomic beam single pass experiments, is the extremely long interaction time of at least several seconds. In this sense an ion storage ring is closely related to ion traps, but with the striking difference of mean relativistic ion velocities. Due to the kinetic transformation and the long interaction time of the stored ions with the laser even such high velocities may be changed significantly by the light pressure force, although the photon momentum is only $5 \cdot 10^{-9}$ of the ion momentum. However, due to the complex motion of the ions, the mean spontaneous light pressure force (eq. 5.11) is considerably reduced in a storage ring.

- Spatially the interaction of ions with the laser beam is restricted to the straight experimental section. In relation to the total ring circumference this gives a duty cycle of $\eta = 0.09$ for the TSR.
- The size of the ion beam in the interaction zone, that is the beam envelope $E(s)$, has been roughly estimated to $3.6 \times 2.2 \text{ cm}^2$. Compared to the laser beam diameter of about 10 mm the geometrical overlap is only $\zeta = 0.1$. However, here a rectangular distribution of the ions within the beam envelope has been assumed, which might not be perfectly valid, so that ζ would be closer to 1.
- Finally the frequency overlap of the absorption line of the stored ions and the laser radiation has to be taken into account. In section 2.3 the modulation of the velocity component v_z due to the betatron oscillations has been derived (2.11). The corresponding line broadening due to the Doppler effect (3.5) yields widths $\delta\nu_B$ in the order of 100 times the natural width $\delta\nu_{nat}$ of the cooling transition (see sec. 3.1).

In summary the mean effective force is

$$\langle \vec{F}_{sp} \rangle = \eta \zeta \frac{\delta\nu_{nat}}{\delta\nu_B} \vec{F}_{sp} \approx 10^{-4} \cdot \vec{F}_{sp} \quad . \quad (5.14)$$

As a consequence for laser cooling of a stored ion beam we indeed need relatively long cooling times to perform the frequency sweep for the Doppler cooling process. Scaling from atomic beam experiments, where velocity profiles of equivalent Doppler widths are cooled within a few milliseconds, we estimate the cooling times to be in the order of 1 s.

If after cooling, the shifted and compressed velocity profile of the ion beam is probed by means of laser induced fluorescence, the relative width of the optical signal $\frac{\delta\nu}{\nu}$ is directly correlated to the momentum spread of the ion ensemble $\frac{\delta p}{p}$. From 3.8 and using $\frac{\delta p}{p} = \gamma^2 \frac{\delta\beta}{\beta}$ one finds

$$\frac{\delta p}{p} = \frac{1}{\beta} \frac{\delta\nu}{\nu_{\pm}} = \frac{1}{\gamma\beta(1 \pm \beta)} \frac{\delta\nu}{\nu_0} . \quad (5.15)$$

For such a measurement it is desirable to work with small intensity of the probing laser beam and preferably in a section of the storage ring, which is separated from the laser cooling section. This is necessary, because otherwise the high fluorescence due to the laser cooling process itself reduces the sensitivity of the detection drastically. The probing section should be free from strongly varying fields and thus betafunctions, as we have seen that this introduces substantial line broadening via longitudinal velocity changes.

5.4 Cooling Limits

Neglecting all heating mechanisms in the storage ring, the theoretical limit for temperatures achievable by spontaneous light pressure cooling is given by the Doppler limit [Leto81]

$$k_B T = \frac{\hbar \Gamma}{2} \quad , \quad (5.16)$$

determined by the natural linewidth of the cooling transition. For the $^3S_1 \rightarrow ^3P_2$ transition we use in $^7\text{Li}^+$, the corresponding temperature is $89 \mu\text{K}$, which is compared to some other ions in table 3.2. In atomic beam cooling experiments using 'optical molasses' for damping the motion of atoms also the dipole force is present and temperatures well below the Doppler limit have been observed [Lett88]. In this case, where the spontaneous emission and thereby the natural linewidth is no longer important, a lower threshold, the recoil limit, may be reached. Here the final temperature is defined by the recoil energy R of an ion due to one single photon recoil.

$$k_B T = R = \frac{\hbar^2 k^2}{2M} \quad (5.17)$$

The extremely low values for this temperature limit are also included in table 3.2. Using a quantum-mechanical coherent superposition of three electronic eigenstates in the He atom it has been possible to circumvent any cooling limit and to reach even below the recoil energy by coherent population trapping [Aspe88]. Although this is rather a passive velocity trapping than an active laser cooling, it impressively shows the perspectives of the laser cooling technique.

In an ion storage ring, on the other hand, several influences act on the ion beam leading to a heating of the stored ion ensemble. These heating mechanisms are counteracting every cooling process and will actually limit the minimum ion temperature to achieve. Describing the statistical evolution of the ion's velocity distribution by the Fokker - Planck equation formalism (see e.g. [Leto81]), the various corresponding heating sources are included in terms of diffusion constants D , defined via the growth of the second moment of the velocity distribution in a time Δt .

$$M^2 < (\Delta v)^2 > = 2 \cdot D \cdot \Delta t \quad (5.18)$$

The influence of each of these effects can be deduced from particular measurements or can at least be estimated from the experimental conditions.

1. Small angle scattering by residual gas molecules leads to a straggling and a growth in momentum spread. This straggling is calculated within the theory of Lindhard et al. [Lind63, Sigm74] and the measurement of broadening of the fluorescence line (see section 4.2) yields a diffusion constant of

$$\frac{D}{M} = 100 \text{ eV/s} \quad (5.19)$$

It turns out, that this is a substantial contribution to the total momentum spread and very clean vacuum conditions have to be required

2. Intrabeam scattering introduces a momentum exchange between ions in the storage ring by Coulomb collisions. Transverse and longitudinal degrees of freedom are coupled, leading to a relaxation of an anisotropic velocity distribution in the rest frame. In principle this yields the possibility to compress also the transverse motion of the ions in a storage ring by laser cooling. Via collisional exchange of kinematic properties between ions it might even lead to a 'sympathetic' cooling of the 1S_0 ground state ions in the beam as observed on Mg-ions in a trap [Lars86]. In the case of $^7\text{Li}^+$ under the experimental conditions of this work, however, intrabeam scattering can be neglected (see section 4.2).
3. Magnetic and electric imperfections are unavoidable at the entrance and exit of focusing elements and the voltage scanning tube electrodes. These lead to small disturbances of the theoretical ion motion, which in case of rational values of the tune Q of the storage ring, cause the beam to be lost immediately (see section 2.3).
4. Photon diffusion heating appears due to the random spontaneous reemission of photons. Assuming resonant laser excitation the momentum change is given by the mean spontaneous photon momentum transfer in a time interval Δt

$$M^2 \langle (\Delta v)^2 \rangle = \langle \hbar^2 k^2 \Gamma \Delta t \rangle . \quad (5.20)$$

Longitudinal cooling of an ion beam thus leads to diffusion constants given by [Puse76]

$$D_{\parallel} = \frac{\hbar^2 k^2 \Gamma}{2} \frac{S}{1+S} (1 + \alpha) \quad (5.21)$$

$$D_{\perp} = \frac{\hbar^2 k^2 \Gamma}{4} \frac{S}{1+S} (1 - \alpha) \quad (5.22)$$

where the parameter $\alpha = 1/3$ takes the isotropy of the spontaneous emission into account. The corresponding frequency spread depends on the interaction time τ_c and the actual scattering rate of photons $\dot{N}$

$$\delta\nu_{diff} = \sqrt{\dot{N}\tau_c} \cdot \Delta\nu_\gamma \quad . \quad (5.23)$$

We estimate the size of the effect to $\delta\nu_{diff} = 3$ MHz for the experimental conditions described below, because at high saturation intensities ($S=50$), the ions being cooled stay out of resonance most of the time. This frequency spread is in the order of the natural linewidth, and thus diffusion heating can be neglected.

Chapter 6

Laser Cooling Experiments

Realising laser cooling of a fast stored ion beam experimentally, demands high laser power in the cooling transition, because the spontaneous cooling force is reduced considerably compared to the theoretical optimum (see section 5.3). Working with one strong laser at about 50 times the saturation intensity of the optical transition, three clear signatures of photon momentum transfer to the ions are observed, as already discussed previously (see sec. 4.2).

1. Scanning the Doppler profile of the stored ion beam in the cooling direction gives an asymmetry of the line shape due to the continuous light pressure shifting of resonant ions.
2. The line center is shifted in addition to the residual gas effect by the laser interaction. This shift also depends on the scanning direction of the frequency.
3. At fixed laser frequency we observe not only the slow decay of the fluorescence due to the storage lifetime, but also a fast decay component within the first 200 ms of laser interaction. This fast decay depends on the laser power and is explained as resonance detuning by multiple transfer of photon momenta.

The first real laser cooling signals, similar to the results of early atomic beam cooling experiments [Andr81, Phil82], have been obtained by the so-called nonresonant cooling technique [Phil85] with voltage tuned probing [Hube89b]. However, at this time the positional and angular control of the ion beam, as well as of the laser beam, has been fairly limited. It is only given within the acceptance of the TSR for the ion beam and by the 35 mm entrance windows for the laser beam.

6.1 Nonresonant Cooling

The ion beam is irradiated by one counterpropagating laser beam tuned to the center of the Doppler profile. Due to the betatron oscillation broadening (see section 5.3) and the high laser intensity ($S \approx 50$), ions within a wide velocity range are decelerated and shifted in frequency. On the low energy side of the laser frequency position the accumulated ions are collected in a narrow peak of a width, in principle determined by the saturated linewidth of the optical transition.

Before the analyzing voltage scan is performed, the laser frequency can be slowly swept across the ion's velocity profile in order to collect them all within a narrow velocity range, but such an integrating effect could not be proved without doubts in this first experiment. The complete time scheme is shown in figure 6.1a. The fluorescence spectrum (6.1b) is taken during the voltage scan, when the laser frequency is fixed. In this situation the ions exhibit constant frequency laser radiation outside the two electrodes, while inside they are Doppler tuned by the applied high voltage of $-5 \dots + 5$ kV. By this scanning voltage the ion's velocity distribution is probed, while the cooled velocity profile is stabilized by the laser standing on resonance outside the tubes.

The compressed signal is enhanced, compared to the uncooled velocity distribution. It contains 50% of the signal integral and its width is 460 MHz detected at PM2 looking inside an electrode (see 4.1). The corresponding beam temperature may be evaluated via 5.2 to 15 K. However, the measurement is distorted by stray field effects from the 200 mm diameter tube electrode [Petr89], which may be seen from the discrepancy of the 730 MHz width of the fluorescence signal detected simultaneously at PM1 shortly behind the electrodes. It will turn out, that also the geometrical overlap of ion and laser beam has not been optimal for these experiments.

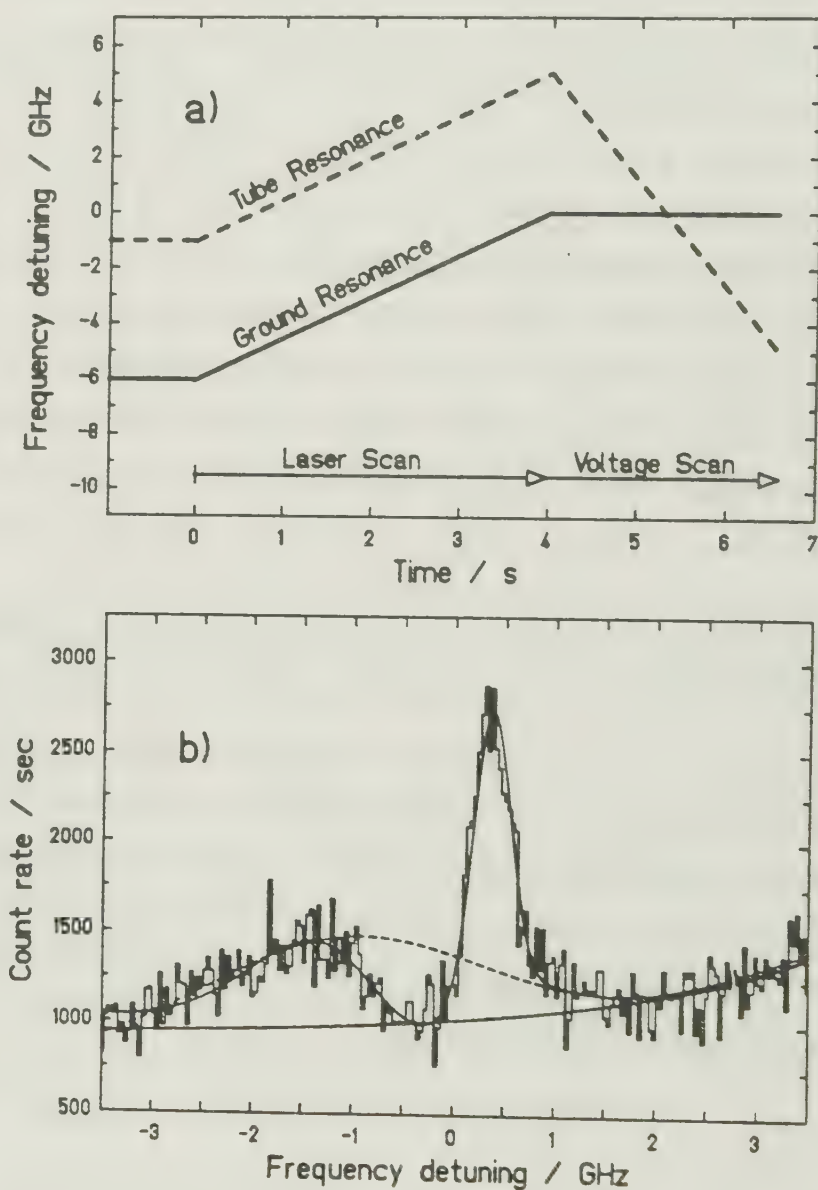


Figure 6.1: Time scheme (a) and fluorescence signal (b) of laser cooling. The spectrum is taken during the voltage scan, while the laser frequency is fixed and is resonant with ions at zero detuning outside the tube electrodes.

6.2 Laser Cooling with two Lasers

When the magnetic rigidity and thereby the kinetic energy of the stored ions could be raised to 1.39 Tm and 13.34 MeV respectively, it has become possible to introduce a second strong cooling force opposite to that of the counterpropagating dye laser beam. The exact ion energy can be matched to an accuracy of 3 keV to a value, where the Doppler shifted transition frequency ν_+ coincides with the 514.5 nm line of an Ar^+ ion laser. When enclosed between these two light pressure forces, the cooled and shifted ion velocity profile is stabilized and the dynamical cooling during the frequency sweep may be extended by a second cooling period. At fixed laser frequencies the ions are trapped in velocity space between the two laser frequency positions and the ensemble will be compressed further. The corresponding acceleration due to two opposing intense laser beams is shown in fig 6.2 a) and b) for different laser frequency separations versus the frequency detuning. The situation in a storage ring, characterized by the reduction of the mean acceleration and the frequency broadening effect, is plotted in the same figure as c) and d). Under these experimental conditions in the moving frame a situation similar to the 'optical molasses' is established, as it is known from atomic beam cooling and trapping experiments [Chu85]. However, again the peculiarities of a storage ring render the application of these results more difficult.

The modified experimental setup (compare sec. 4.1) is shown in figure 6.3. The single-mode Ar^+ ion laser 1 can be stabilized to a confocal Fabry Perot interferometer to a short time stability < 1 MHz [Grie89]. For applications to precision experiments the stabilization to the $^{127}I_2$ secondary wavelength standard is prepared [Krie89].

The laser cooling procedure is now like the following. The frequency of the copropagating single-mode Ar^+ ion laser beam is kept constant and is set in resonance with ions at the low energy side of the velocity profile of the ion beam. The frequency of the counterpropagating tunable dye laser beam is correspondingly set to the high energy side. The combined light pressure force of both lasers is discussed in frequency space in section 5.3. When sweeping the latter frequency towards higher values, ions getting into resonance are decelerated and accumulated in a narrow peak. The probing of the cooled velocity profile of the ion beam is done in different ways.

1. The fluorescence is detected during the cooling laser sweep. When the scanning laser frequency is swept across the Ar^+ ion laser frequency in the ion's rest frame,

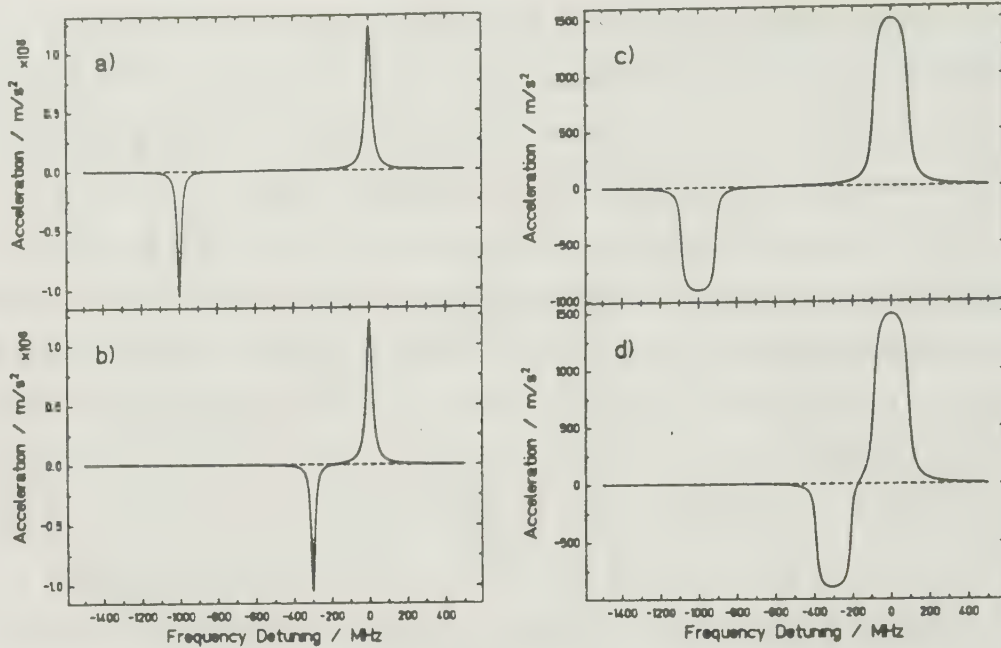


Figure 6.2: Effective light pressure acceleration in the field of two oppositely directed laser beams of 900 and 400 mW/cm^2 with frequency detunings of 1000 and 300 MHz in the ion's rest frame. a) and b) show the ideal situation, which is modified in a storage ring due to the betatron oscillations and the duty cycle (c) and d)). An induced betatron width of $\delta\nu_B = 180$ MHz is assumed (compare sec. 3.1).

the cooled ions are brought to bright fluorescence in the combined field of both co- and counterpropagating laser beams.

2. Stopping the laser frequency sweep before this crossing point, the accumulated ions are 'trapped' in velocity space between the two strong longitudinal light pressure forces. In contrast to the dynamical procedure described above in this steady situation the enclosed ensemble is further cooled down until an equilibrium between the cooling forces and the heating mechanisms in the ring is reached. The probing is done by a high voltage scan applied to the tube electrodes.
3. An independent measurement of the beam properties of all ions (1S_0 ground state and all 3S_1 metastable) is achieved with the electronic Schottky noise signal (see section 6.5). The Schottky spectra are also taken after the laser cooling sweep has stopped, enclosing the cooled ions between the two laser frequencies.

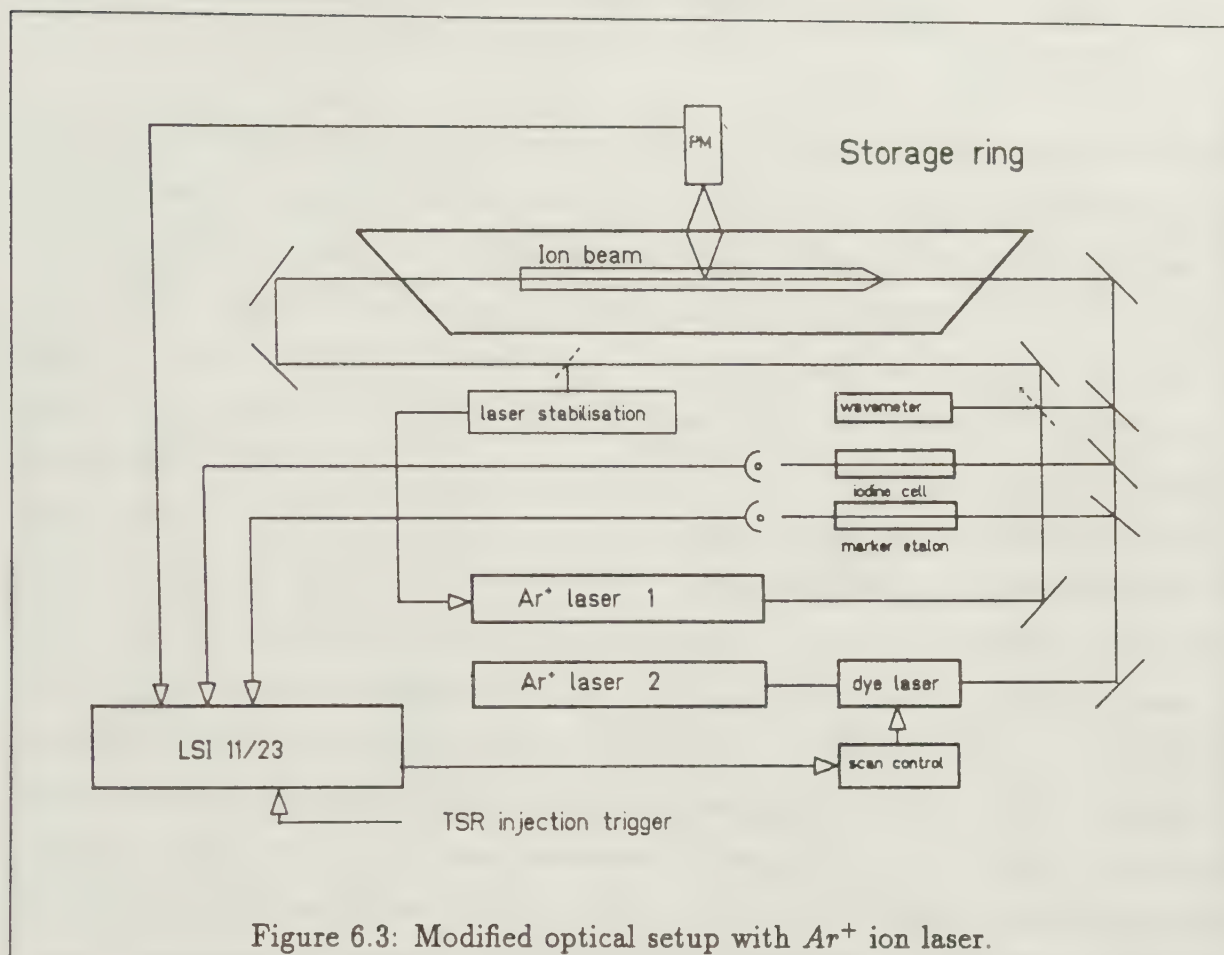


Figure 6.3: Modified optical setup with Ar^+ ion laser.

4. An independent fluorescence measurement of the ion's velocity distribution by a weak probe beam in a separate section of the TSR could not be performed up to now due to technical reasons. However, this would be the appropriate method for detection of the achieved ion temperature.

6.3 Laser Cooling with Limited Geometrical Beam Control

The laser cooling efficiency depends naturally on the laser intensity, scanning speed, and the experimental geometry. Typical fluorescence signals, taken before optimization of the geometrical overlap and including the crossing of the laser frequencies in the ion's rest frame are shown in figure 6.4. The series of spectra clearly shows the dependence of the position of the cooled peak from the Ar^+ ion laser frequency, variable within ~ 6 GHz under the Ar^+ Doppler profile. In these measurements the absolute frequency is only known to an accuracy of ± 300 MHz, limited by the commercial wavemeter [Bur85], but the relative stability even without active stabilization is below ± 20 MHz. The question of collection efficiency of the laser cooling cycle is answered by voltage scanning measurements. The cooling laser sweep is stopped before the point is reached, where an ion can be resonant with both lasers simultaneously, and, with a variable delay in between, the analyzing voltage scan probes the cooled as well as the original velocity distribution of the beam (see fig. 6.5a). Independently of the systematic errors of these voltage scans the signal in figure 6.5b, which shows fluorescence spectra taken during this scan with and without laser cooling, proves almost 100% of the metastable ions in the beam to be cooled and accumulated in the narrow peak.

A second question to be answered by these measurements is the lifetime of the cooled ions in the TSR. In appendix D similar measurements with different delays are shown, which yield the same lifetime of 1.7 sec for the enclosed cooled ions as the total 3S_1 lifetime has been in this series of measurements.

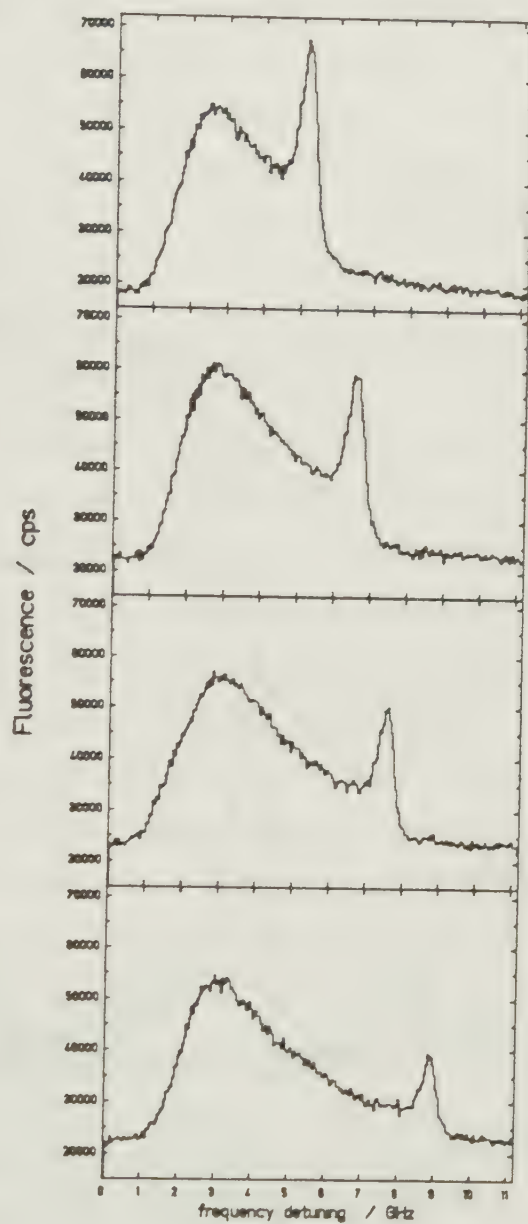


Figure 6.4: Laser cooling fluorescence spectra with different positions of the Ar^+ ion laser frequency

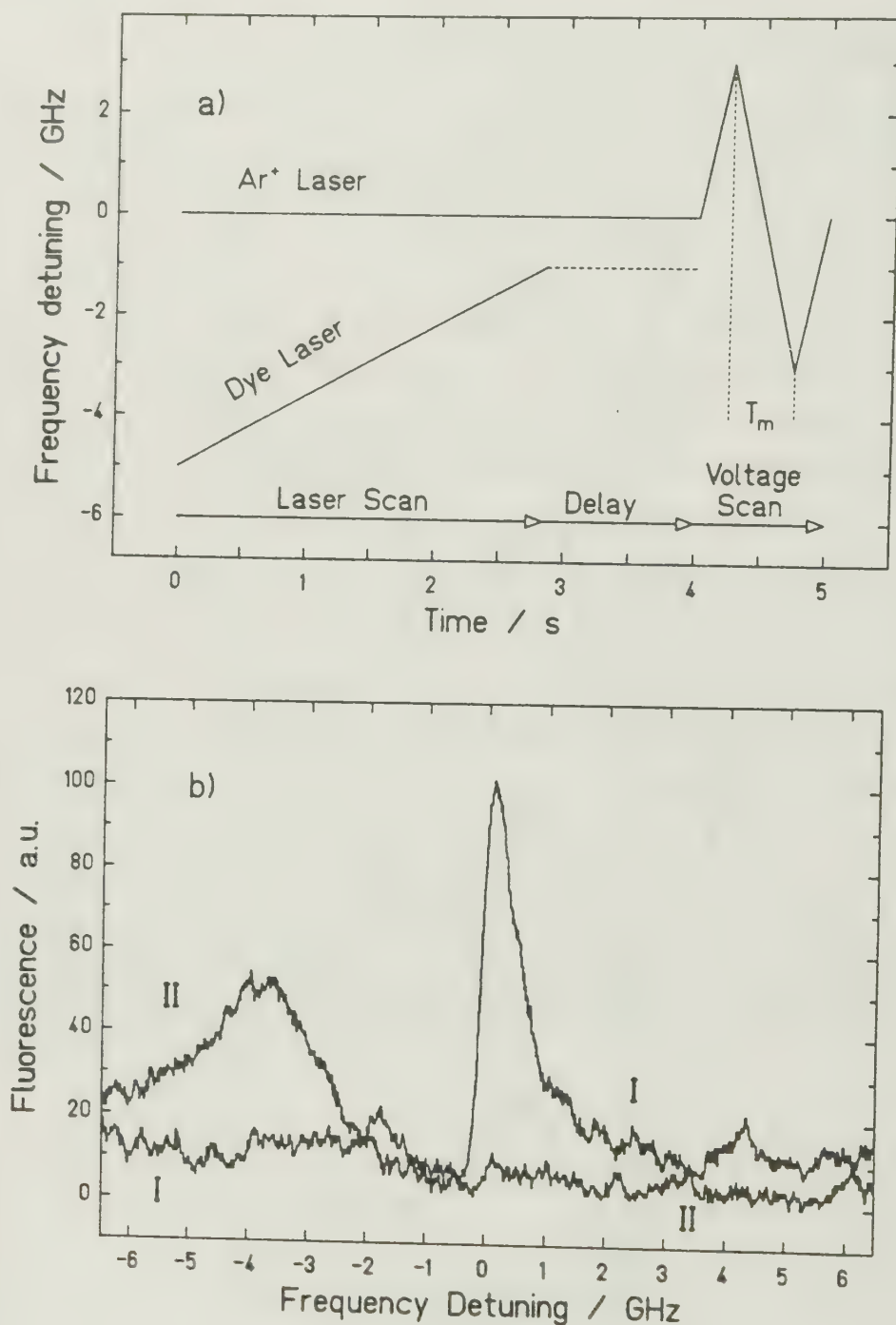


Figure 6.5: For probing the ion's velocity distribution Doppler tuning is applied. A slow cooling laser sweep is followed (after a variable delay) by the analyzing voltage scan covering the cooled and the original distributions (a). The copropagating Ar^+ ion laser beam is blocked during this probing. (b) shows two spectra taken with (I) and without (II) laser cooling during the time interval T_m .

6.4 Laser Cooling after Geometrical Alignment

Drastically improved control over the experimental geometry at the TSR is achieved by the insertion of three 18 mm apertures into the ring and adjustment of the ion beam through this collimator. The necessity of a totally new adjustment of the ion beam orbit in the TSR shows, that up to now, large angle deviations must have been present between the ion beam and the geometrical TSR axis. With careful adjustment, 25% of the total ion beam can pass through the apertures. By centering the two laser beams of 10 mm diameter through the apertures as well, the geometrical overlap of the ion beam with the two laser beams is improved considerably, defining the angle between both to $\Theta \leq 1 \cdot 10^{-3} \text{ rad}$. Switching now between operation with and without this collimator does not change the geometry, but allows for studying situations with more or less betatron oscillation amplitudes.

This adjustment has influenced the laser cooling performance dramatically. The more effective collection of ions may be seen from the strongly pronounced tail of the Doppler profile to the high frequency side. The signals shown in figure 6.6 are measured with the collimating apertures inserted into the TSR. The original distribution seen in trace (a) has been obtained by reverse scanning (decreasing laser frequency), thus avoiding the cooling compression. Normal scanning leads to 'Doppler cooling' and velocity compression as shown in spectrum (b). While scanning over the initial distribution nearly all ions in the metastable $^3S_1(F = 5/2)$ state are collected. The constantly increasing fluorescence in this range reflects the integrating character of the ion collection process. Ions shifted out of the original velocity distribution are continuously scattering photons resulting in an almost constant fluorescence intensity until finally the cooled ions are detected by scanning the dye laser frequency across the Ar^+ ion laser frequency in the ion's rest frame. The apertures are mechanically cutting the ion beam diameter to 18 mm and thereby the maximum betatron amplitudes of the ions are also restricted to this value. By removing the apertures the influence of ions with large oscillations may be studied without changing the geometrical alignment of ion beam and laser beams. In order to fix this difference qualitatively, series of laser cooling spectra are measured with varying scanning speeds of the cooling laser frequency and are shown in figure 6.7. The evaluated dependencies of the area and width of the cooled peak from the scanning speed are also contained in figure 6.8. The area, corrected by the storage time, is plotted in arbitrary

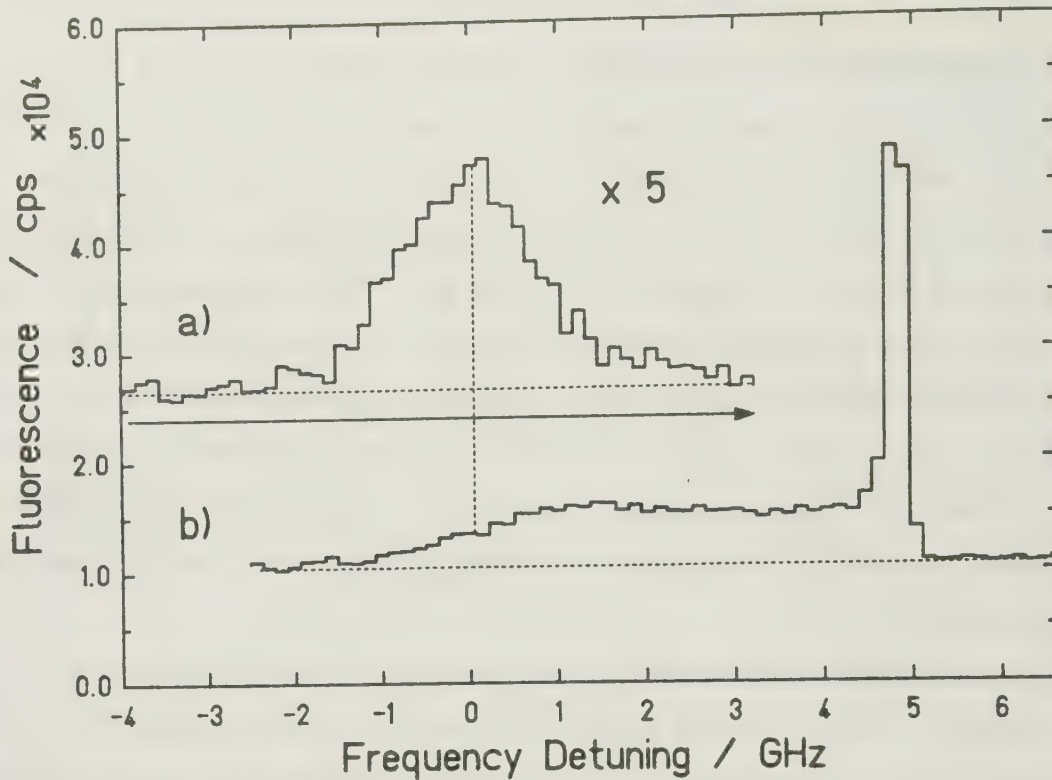


Figure 6.6: With the apertures inserted into the TSR the ion beam Doppler profile is scanned in different direction. Reverse scanning yields the original velocity distribution (a), while scanning the cooling direction shows the integration character of the laser cooling procedure (b).

units, but the normalization of data with and without the apertures can only be done roughly by comparison of general fluorescence count rates in both cases. While without the apertures the accumulated number of ions, assumed proportional to the peak area, is constantly increasing with the scanning speed, in the case of cutting the betatron oscillations it saturates at about 4 GHz/s. This cooling speed differs from atomic beam cooling experiments and estimates from the maximum spontaneous light pressure force (5.11) by a factor of $\approx 10^3$, which is in agreement with the reduction of the interaction time as measured by the optical pumping experiment in section 4.2.

The width of the cooled peak does not depend systematically on the scanning speed, but differs slightly in the two series of spectra. Due to the nonlinear dependence of the fluorescence yield in the very simple probing technique of shifting the ions across the Ar^+ ion laser frequency, however, only an upper limit for the temperature of the cooled

ensemble may be quoted from these measurements. The average width at half maximum of 200 MHz corresponds to a beam temperature (via eq. 5.2) of

$$T \leq 3K \quad . \quad (6.1)$$

Besides the cooling itself, an additional quality of the laser cooling technique lies in the extreme precision of defining the energy of the ion beam. The achieved relative energy width of $\delta E/E = 4 \cdot 10^{-11}$ yields a high potential, e.g. for high voltage calibration [Poul82] and in many other applications. The absolute energy change of the ions by laser deceleration is larger than 5.5 keV in the laboratory frame. This number is principally not limited, except by technical means due to laser scanning range limitations. In this specific series of measurements the restriction is given by the shifting of the Ar^+ ion laser frequency under the Ar^+ Doppler profile. As an important point, however, it has been proved, that ion beams with initially very much larger momentum spreads can also be cooled efficiently by this technique. Beams with $\Delta p/p$ up to the 10^{-3} region, as expected at the ESR, may be compressed within the same cooling time of a few seconds to equivalent longitudinal temperatures.

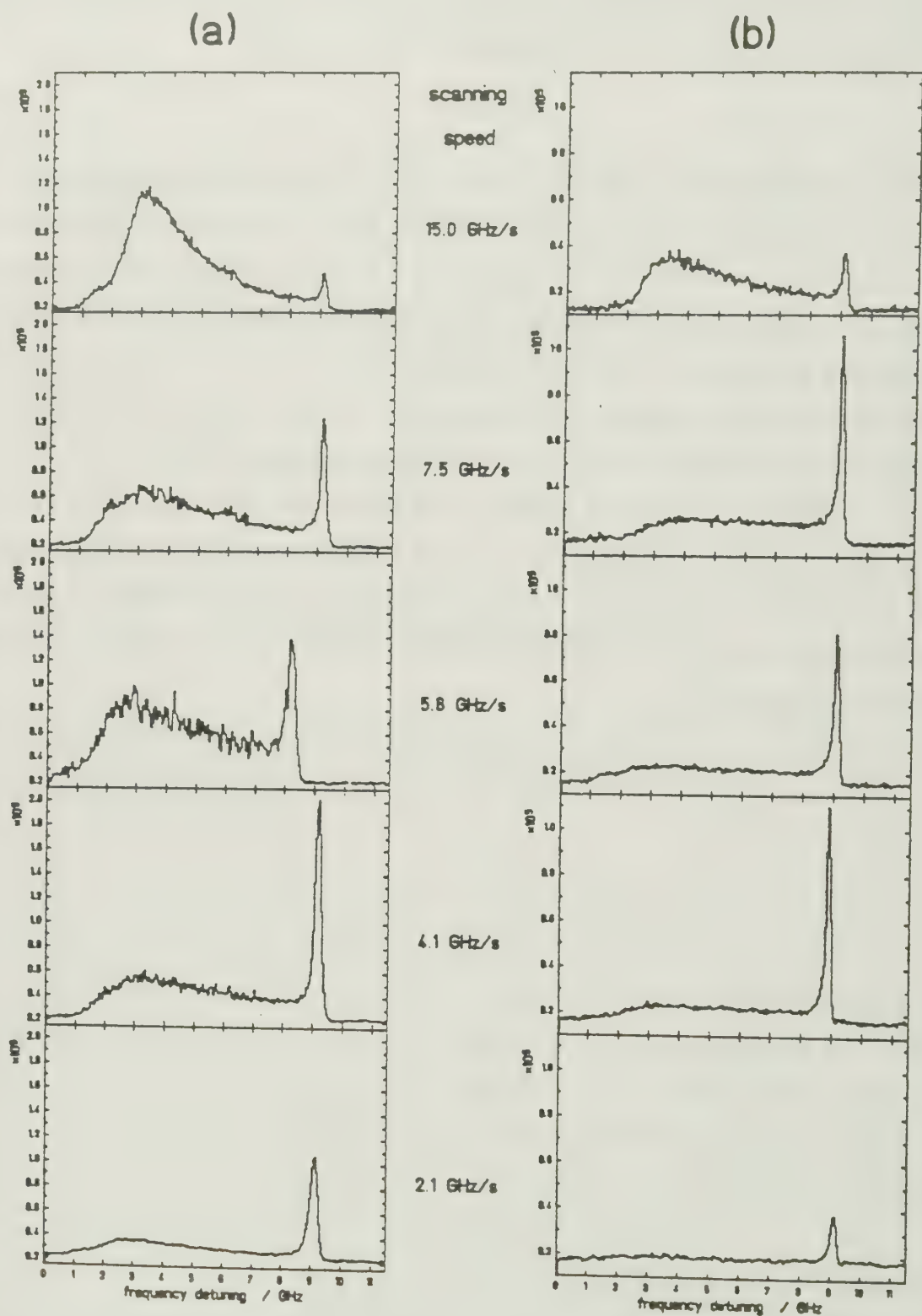


Figure 6.7: Fluorescence spectra of laser cooling are shown for different scanning speeds of the cooling laser frequency. The fluorescence yield is plotted in units of $10^5/s$. The spectra are not corrected for the storage time of the ion beam. The scanning speed of each two corresponding spectra is given between the series (a) and (b), taken without and with the apertures inserted into the TSR.

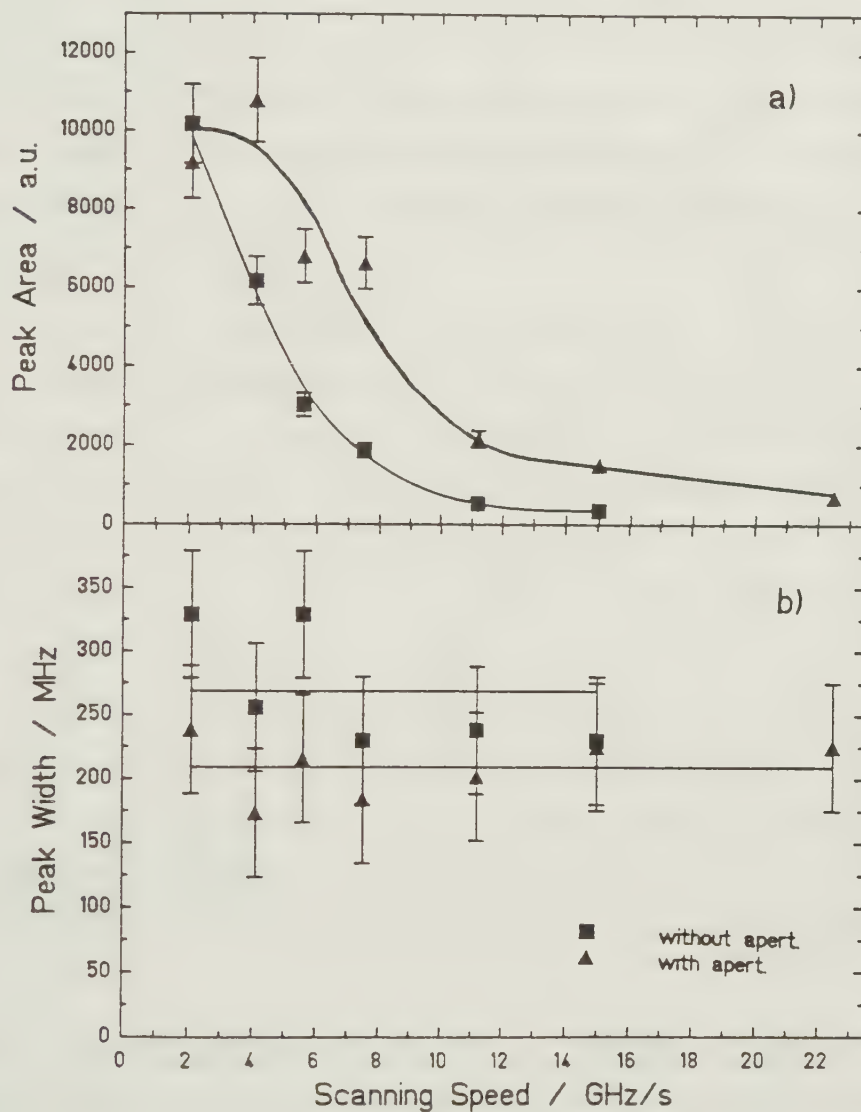
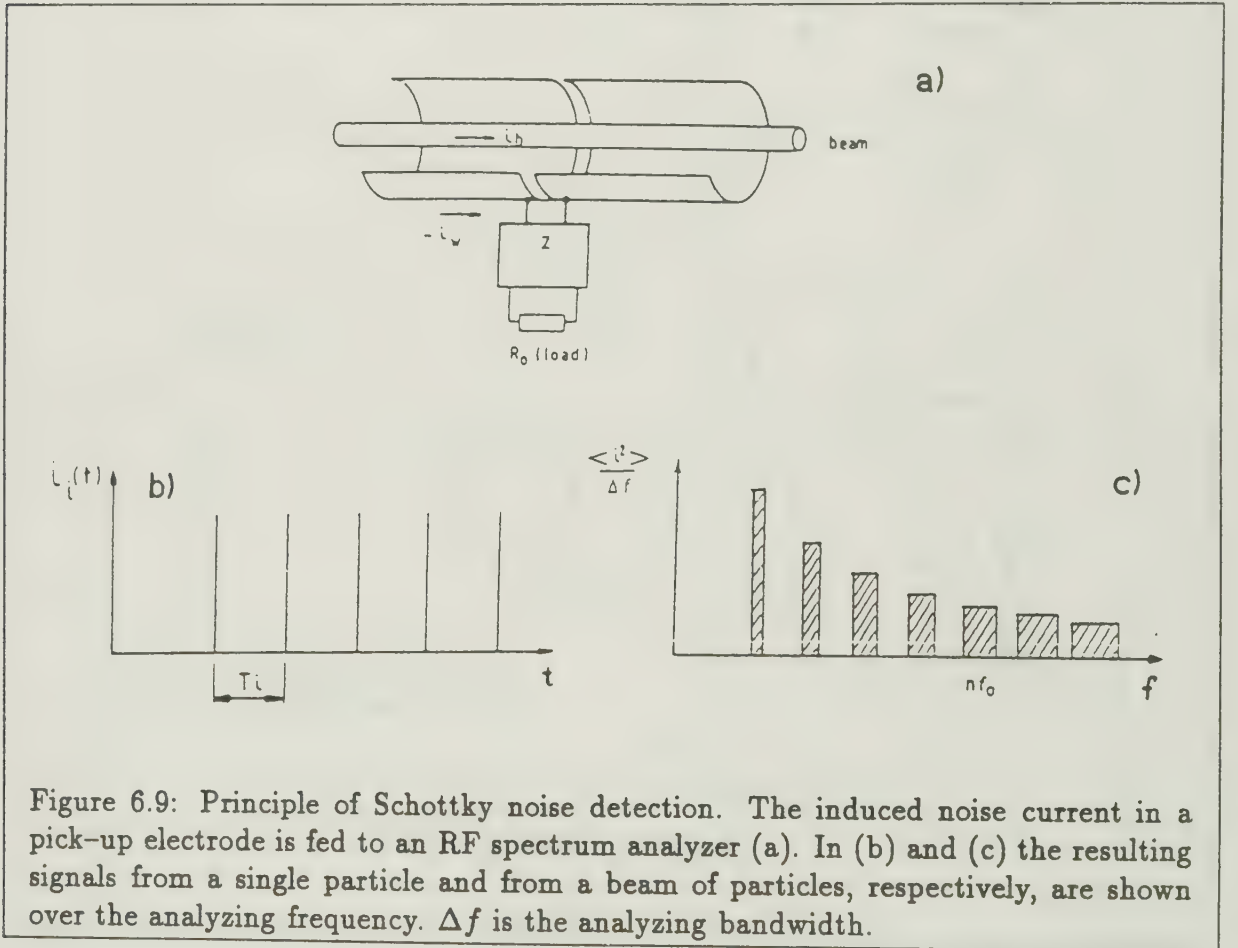


Figure 6.8: Results from figure 6.7. The dependence of the area (a) and width (b) of the laser cooled narrow peak from the scanning speed is plotted for both cases with and without the apertures inserted. The data are corrected for the storage time of the beam.

6.5 Schottky Noise Signals

Independent from the fluorescence detection, ion beam properties can be measured by the Schottky noise signal, the standard diagnostic tool at an ion storage ring [Tur85, Chat85]. The electronic noise of the stored ion beam is detected by a longitudinal pick-up electrode and the frequency distribution is obtained by an RF spectrum analyzer. The principle of operation may be seen from a schematic view in figure 6.9. An ion passing the electrode induces a current $i(t)$, which for a single particle with revolution frequency f_i would give an infinite chain of delta pulses in the time domain. In the frequency picture this corresponds to a spectrum containing all harmonics $n \cdot f_i$. For N particles, randomly



distributed around the storage ring with small deviations from the mean momentum p_0 (of width δp), each delta function at $n \cdot f_i$ in the Schottky spectrum is replaced by a band of frequencies of relative width

$$\frac{\delta f}{n \cdot f_0} = \eta \cdot \frac{\delta p}{p_0} \quad (6.2)$$

f_0 is the mean revolution frequency of all ions and η is the machine parameter defined in 2.3. This relation is valid for hot, low-density, continuous ion beams, and in this case the square of the measured signal at the spectrum analyzer $|A_n(f)|^2$ is proportional to the revolution frequency distribution of the stored ions. The integral $\int |A_n(f)|^2 df$ is proportional to N . For cooled beams, additional peaks may occur in the power spectrum at phase velocities, where collective fluctuations can propagate as waves [Park80].

With the apertures inserted, figure 6.10 shows a laser cooling measurement, where the cooling laser sweep is stopped before reaching the point where ions can be resonant with both lasers (compare appendix E). The amplitude signals are measured within several seconds, delayed by 2.5 s after ion injection, the time needed for the laser cooling sweep. Trace a) shows the original frequency width without any laser interaction, while spectrum b) has been recorded after laser cooling at laser intensities of the co- and counterpropagating beams corresponding to saturation parameters of $S = 130$ and $S = 50$ respectively. The broad distributions in a) and b) due to the uncooled ions show equal widths and power integrals, that is, particle numbers. The shifted and compressed metastable $^3S_1(F = 5/2)$ ions cause a narrow resonance at f_{cool} . The signal width is determined by the bandwidth of the spectrum analyzer, set to $\Delta f = 50 \text{ Hz}$ in this case. Within 5% the relative shift of the intense narrow Schottky peak agrees with the frequency shift of the velocity distribution of cooled ions determined by a fluorescence measurement. Due to the high saturation of both lasers the position of the cooled ions is assumed to be in the middle between the two laser frequencies. A high resolution Schottky signal is inserted as trace c). It is taken at the 36th harmonic and adapted to the frequency scale of the previous spectra. The effective bandwidth is as low as 14 Hz and the observed signal width of 30 Hz corresponds to a relative width of $\Delta f/f = 2.5 \cdot 10^{-6}$.

However, this width is probably not directly connected to the actual momentum spread of the cooled ions, because the obvious signal enhancement in the cooled peak indicates the presence of collective effects within the ion beam. If one assumes that the narrow peak is caused by incoherent single particle fluctuations ($|A(f)|^2 \propto N$), the ratio of cooled to uncooled ions is $\frac{N_{cool}}{N_{hot}} = 30$. This value is about 100 times larger than expected from the metastable beam fraction (0.1 – 0.3), deduced from fluorescence signals. From the 100 ms lifetime of Bennett holes produced by optical pumping (see section 4.2), any collisional cooling of ground state ions by the laser cooled metastable ions is ruled out due to low exchange rates. Therefore only a strong signal enhancement, produced by a

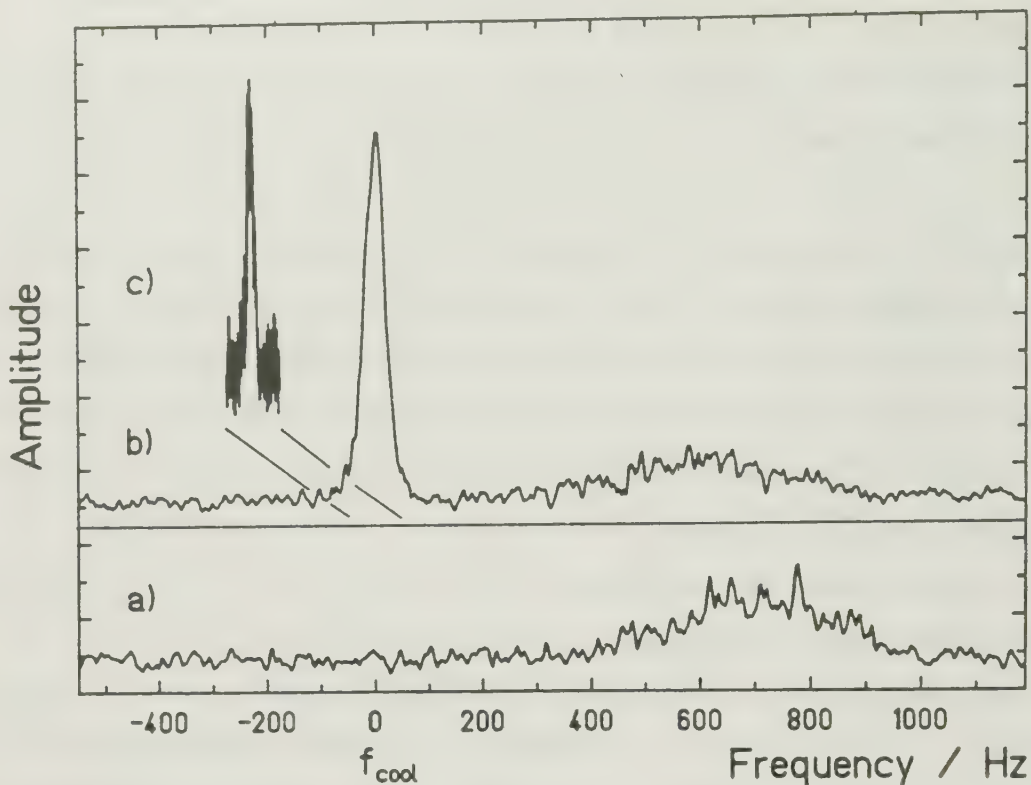


Figure 6.10: Schottky noise signals. The signals are taken with a delay of 2.5 s after the injection of the ions into the TSR, the time required for the laser cooling sweep. They are shown in a linear scale. Traces (a) and (b) are taken with identical sensitivity but different resolution bandwidth of 100 Hz and 50 Hz, respectively, at the 18th harmonic of the revolution frequency at $f_{cool} = 6.210340$ MHz. Without laser cooling spectrum (a) is obtained, and performing the laser cooling sweep yields spectrum (b). Finally trace (c) – adopted to the same frequency scale – is taken at the 36th harmonic resulting in a higher frequency resolution.

collective beam response, can explain the observed spectra. In addition to this, also the integral power spectrum is by a factor of 4 larger than the corresponding integral of the uncooled spectrum. This is a totally new phenomenon in an ion storage ring, which we observe for a large span of harmonic numbers ($6 \leq n \leq 36$), different laser intensities ($1 \leq S \leq 100$), and different energy shifts between cooled and uncooled distributions.

From electron cooling experiments on high intensity ion beams, distortions of the shape of the Schottky spectrum due to collective waves inside the stored ion beam are well known [Park80, Poth89, Stec90] (see appendix F). These waves, travelling along with and opposite to the mean ion velocity, lead to two narrow peaks at the equivalent phase

velocities in the Schottky spectrum. Simultaneously the integral noise power is reduced considerably. This noise suppression is due to screening effects in cooled and intense ion beams, that is at high ion densities.

In our case only $2 \cdot 10^7$ metastable ${}^7\text{Li}^+$ ions are injected into the TSR and their number still decreases due to the limited storage time by about a factor of 5 until the Schottky spectra are taken. In order to describe the stability conditions of a stored ion beam, regarded as a one component plasma, the longitudinal coupling impedance $Z_{||}$ is introduced as a new machine parameter. The main properties of the beam's electromagnetic field and the influence of the physical beam tube of the storage ring are included in $Z_{||}$ [Chat85, Bry85]. Four major components contribute:

- a resistive wall component at very low frequencies,
- narrow band resonances, introduced e.g. by the RF cavity,
- a broad band impedance due to numerous sudden cross-section variations along the ring circumference, and
- the space charge component.

The parameter $Z_{||}$ is specific for any particular storage ring and the threshold for ion beam instabilities to show up, is given in terms of the particle number N by the longitudinal beam temperature $T_{||}$ and $Z_{||}$ according to the Keil-Schnell criterion [Keil84, Bry85, Jaes89].

$$N > N_{crit} = \frac{2T_{||}}{q^2 e^2 f_0} \cdot \left[\frac{\text{Im}(Z_{||n})}{n} \right]^{-1} \quad (6.3)$$

$$= 3.125 \cdot 10^9 \cdot \frac{nT_{||}/K}{\text{Im}(Z_{||n}/\Omega)} \quad \text{for our experimental conditions,} \quad (6.4)$$

where q and f_0 are the charge state and revolution frequency of the beam and n is the harmonic number of the Schottky analysis. For a typical impedance of the TSR of $1k\Omega$ this criterion demands $N > 1 \cdot 10^7$ ${}^7\text{Li}^+$ ions at 3 K temperature for plasma instabilities to develop, and we are slightly below this critical value under the present experimental conditions.

The ion density is here considerably lower than in the electron cooling experiments mentioned above, but there are two other important features which distinguish the situation of a laser cooled ${}^7\text{Li}^+$ ion beam from electron cooling. Firstly the laser cooling force

acts on a narrower velocity range, the force itself and the gradients in velocity space are much larger than in electron cooling. Secondly the laser cooling sweep shifts part of the stored ions out of the original velocity distribution, while during electron cooling the one peak structure of this distribution is conserved. In this specific situation the existence of two stream instabilities is well known in plasma physics [Laws77]. However, also for this effect the Keil-Schnell criterion should hold, and it is not clear up to now, what is the reason for the observed signal enhancement.

In a series of Schottky noise measurements of the laser cooled ion beam the intensities of the cooling laser as well as of the Ar^+ ion laser are decreased drastically. In figure 6.11 it can be seen that the narrow peak in the Schottky spectrum is detected down to saturation parameters of $S_{dye}=0.3$ and $S_{Ar^+}=0.7$. Decreasing the intensities further the peak disappeared abruptly. Unfortunately the fluorescence signal is too weak at these conditions so that no information about the cooling efficiency is available from these data. Nevertheless, the limit of $S \approx 1$ for the laser cooling effect to be present is in agreement with the saturating behaviour of the spontaneous light pressure force (eq. 5.11), which quickly drops to zero for $S < 1$.

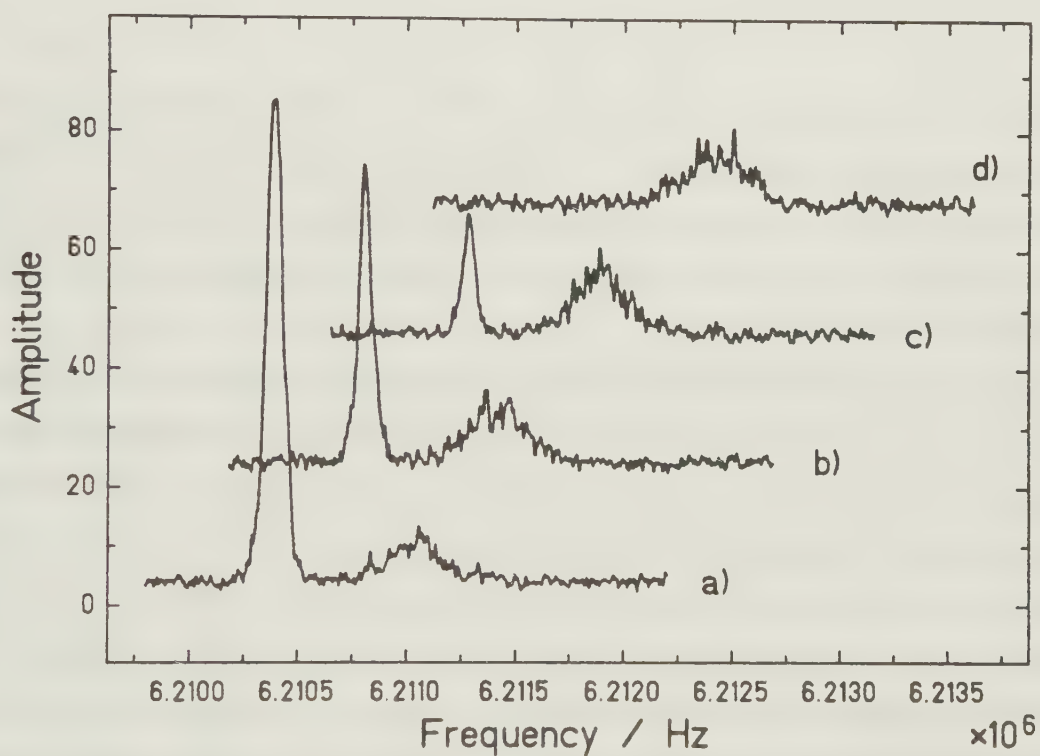


Figure 6.11: Schottky signals taken after laser cooling at different laser intensities: (a) $S_{dye}=40$, $S_{Ar+}=130$; (b) $S_{dye}=10$, $S_{Ar+}=5.8$; (c) $S_{dye}=0.3$, $S_{Ar+}=0.7$; (d) $S_{dye}=0$, $S_{Ar+}=7.2$. The timing is identical with 2.5 s delay and 2 s sweeping of the spectrum analyser.

Chapter 7

Conclusion

Due to the rapid technical development of the TSR the measurements described in this thesis have been performed at different operating parameters and settings of the machine. Nevertheless, many important questions accessible to laser spectroscopy are investigated and the technique of laser cooling is for the first time applied to a relativistic ion beam in a storage ring.

Laser spectroscopy of the stored ion beam yields detailed information about ion beam properties like momentum spread, storage time, or the influence of residual gas scattering. Dynamic processes like the mixing of velocity classes in the stored beam are investigated and the long time constants of 100 ms for this effect make laser cooling possible. The 10^{-4} reduction of the mean light pressure force due to the specific ion kinematics in a storage ring can be overcome by the long interaction time of several seconds. Although the ${}^7\text{Li}^+$ beam from the tandem accelerator already shows a quite narrow longitudinal momentum spread of $\delta p/p = 6 \cdot 10^{-5}$, it is compressed by at least a factor of 10 by laser cooling to a final value of $6 \cdot 10^{-6}$. The probing of the final velocity distribution has to be improved by means of low intensity excitation of the ions in a section of the TSR separate from the cooling region. From the present measurement we can only quote an upper limit for the longitudinal beam temperature of 3 K.

After laser cooling, collective plasma waves within the stored ion beam are detected in the Schottky noise signal. These instabilities are not finally understood as yet, but represent an interesting effect in a well defined one-dimensional plasma.

Concerning the precision experiments of optical spectroscopy of M1 transitions and saturation spectroscopy in the stored ion beam, our measurements show very much the feasibility of this type of experiments. First Lamb dips in the Doppler broadened fluorescence line are observed. Due to the large transverse motion of the ions the obtained dip

widths are rather large and transverse cooling of the ion beam seems necessary in order to reach the resolution one aims at [Hube87, Klei89].

In contrast to the longitudinal laser cooling described here, stochastic and electron cooling techniques provide such three-dimensional compression of the ion motion. An electron cooling device is implemented in the TSR, but at the present stage a 9.12 MeV ${}^7\text{Li}^+$ beam could not be cooled by the merged electron beam due to various reasons. A test at higher ion energy (13.3 MeV), and with improved geometrical overlap, has not yet been performed. Improved vacuum conditions in addition will prolong the available time for electron cooling. The advantages of both electron cooling and laser cooling, may be connected to high performance ion beam cooling. Such a combined cooling scheme has been considered theoretically by [Wolf88]. The inclusion of transverse cooling by the magnetized electron beam, and the extension of the cooling force to a much wider range in momentum space, are the most striking features.

In atomic beam cooling, great improvements have been made in building up quasi optical elements for controlling the transverse atomic motion as well [Baly85, Nell89], but for stored ion beams only few ideas exist for transverse laser cooling [Chan81, Baly89]. Due to the very short interaction time in such a transverse setup high laser intensities are needed and special techniques like π -puls excitation of the ions must be applied. A second way of transverse laser cooling is to couple the longitudinal and transverse motions of the ions in the storage ring and thereby transfer the longitudinal cooling effect to the other degrees of freedom as well. This may be done, for example, by increasing the ion density in the beam, so that intrabeam scattering becomes effective. The Coulomb collisions in this process lead to an equilibrium temperature of the ion beam in all three dimensions, and after a correspondingly longer cooling time the transverse motion will also be damped. Sympathetic cooling of the large fraction of ${}^1\text{S}_0$ ground state ions is an additional effect of this mixing and thus all ions in the ring will be cooled. A high current ion source as an injector for the Heidelberg TSR is under construction and will give access to this high ion density regime.

The cooling time itself, also only in one dimensional cooling, may be reduced considerably by making use of the strong dipole force component of the light pressure force. As shown theoretically [Dali85], it may be two orders of magnitude larger than the spontaneous force, if the settings are chosen properly. In principle we already work in an appropriate experimental geometry, but very high laser power densities and a clean geo-

metrical situation are needed to involve this force. First experiments observing indications of dipole force action on a fast atomic beam of neutralized 3 keV $^{20}\text{Ne}^+$ ions have shown these difficulties [Jess89].

Although laser spectroscopy of stored ions has turned out to be a very powerful diagnostical tool, giving detailed and accurate information about ion beam properties, it will be extremely helpful for precision spectroscopy as well as for ultimate laser cooling to do time resolved fluorescence spectroscopy. As in ion traps [Died87b] the intensity autocorrelation function

$$G^{(2)}(\tau) = \langle I(t) \cdot I(t - \tau) \rangle \quad (7.1)$$

can reveal information about the detailed oscillational motion of stored ions in the ring. On a time scale of the revolution frequency, the observation of a velocity echo from tagged velocity classes of ions will yield similar information [Hube89a]. In order to compare such measurements with the theoretical motion of ions in a storage ring it is necessary to perform Monte Carlo simulations. A computer code for such single particle tracking is in preparation.

An effective cooling in all three degrees of freedom to temperatures in the order of the Doppler limit ($89\mu\text{K}$) might lead to phase transitions and to the onset of ordering within the ion beam. The Schottky noise signals already indicate ion densities, where collective phenomena like plasma waves and instabilities show up. Theoretically the final stage of an ultimately cooled beam will be crystallization [Hans75, Schi86, Habs86]. The Doppler limit temperature corresponds to a maximum Γ - factor, defined as the ratio of the Coulomb energy $\frac{(Ze)^2}{d}$ between ions to the thermal energy $k_B T$, of

$$\Gamma = \frac{\frac{(Ze)^2}{d}}{k_B T} = 7500 \quad (7.2)$$

This is about forty times the critical value ($\Gamma_{crit} = 170$) required for the formation of crystals, but this is only valid for the longitudinal motion. Transversely the temperature is still in the order of 10^4 K with inserted apertures, and although the calculated characteristic length of $d = 26\mu\text{m}$, the approximate distance between neighbouring $^7\text{Li}^+$ ions of a beam crystal in the TSR, is much larger than the linear particle density of $\approx (0.3\mu\text{m})^{-1}$, this is a strong hinderence for crystallization.

The absolute energy of the cooled ions is defined by the cooling laser frequency. It may be varied within about 20 keV and the energy value can be determined to high accuracy from an optical frequency measurement. From a direct calibration by molecular

$^{127}\text{I}_2$ absorption lines, one gets a precision in ion momentum of $\delta p = 3 \cdot 10^{-6} p$, which may be increased by at least three orders of magnitude, when stabilized lasers are used and locked to optical frequency standards [Spie77, Spie80]. The possibility of shifting the cooled ions far in energy, clearly demonstrates the feasibility of laser cooling ion beams with initially much larger relative momentum spread as well. Beams with $\delta p/p$ up to few times 10^{-3} , as expected at the ESR in Darmstadt, can be cooled effectively to similar temperatures as demonstrated here.

A possible technical application of the deceleration by laser interaction lies in the field of ion injection techniques at storage rings. By the RF stacking method, single buckets of injected ions are slightly decelerated by $\delta p/p \approx 0.02$ by means of an applied RF field in a special section of the TSR. Thus the whole momentum acceptance of the machine is filled subsequently by up to 20 buckets [MPI88]. In principle the same procedure can be performed by laser deceleration avoiding the heating due to the RF field. A momentum reduction of 10^{-3} by the light pressure will give a sufficient horizontal offset of $\approx 2\text{mm}$ at the septum in the injection region (dispersion $D=2\text{m}$). No doubt the time needed for such a 'laser stacking' is much longer than for the RF method, but on the other hand this technique is automatically coupled with the laser cooling effect and the result will be an accumulation of ions in a cold dense beam.

In section 6.5 it is shown that laser cooling even works with very low laser intensities. This fact implies the possibility of applying the laser cooling technique to ions with weaker optical transitions like M1 transitions between fine- or hyperfine levels as well. The list of candidates may be extended drastically, including ions for which the corresponding transition frequencies lie in the optical region – in principle all those proposed for optical precision spectroscopy on M1 transitions like S^{11+} (fs) or hydrogenlike Bi^{82+} (hfs) [Kühl87b, Kühl88]. The transition rates are typically three orders of magnitude lower than for E1 transitions, and consequently the lifetimes of the excited states are in the order of ms.

Looking at storage ring facilities other than the Heidelberg TSR and the Darmstadt ESR, very much higher magnetic rigidities and ion energies are available in principle. At the CERN SPS e.g. at $B\rho = 1000\text{ Tm}$, hydrogenlike $^{16}\text{O}^{7+}$ may be stored at 200 GeV/u . The corresponding relativistic factor is $\gamma = 200$ and due to the Doppler effect the ground state transition $1s \rightarrow 2p$ is shifted into the optical frequency region to about 535 nm . The re-emitted photons in forward direction, on the other hand, will be 185 keV x-ray

photons and the whole scheme may be regarded as a tunable narrow bandwidth x-ray source.

These few examples of new forthcoming ideas demonstrate the various possibilities in this wide open field of experimental physics at ion storage rings.

Appendix A

Saturation Intensity

The saturation intensity I_s of a transition is defined for a two level system with energy spacing $\hbar\omega_0$ by equality of the induced excitation rate and the spontaneous decay rate A of the upper level. Following [Cohe88] the saturation parameter $S = \frac{I}{I_s}$ on resonance is defined as

$$S = \frac{2\Omega^2}{A^2} \quad , \quad (\text{A.1})$$

where $\Omega = -\frac{\vec{d} \cdot \vec{E}}{\hbar}$ is the Rabi frequency with $\vec{d}$ the dipole matrix element and $\vec{E}$ the electric field vector. In the ideal case (*) of $\vec{d} \parallel \vec{E}$ the Einstein coefficient A is given by

$$A = \frac{4\omega_0^3}{3c^2\hbar} d^2 \quad (\text{A.2})$$

and thus

$$S^* = \frac{3E^2 c^3}{2\hbar\omega^3 A} \quad . \quad (\text{A.3})$$

The laser intensity I is proportional to the squared field amplitude

$$I = \frac{c}{4\pi} E^2 \quad (\text{A.4})$$

and for the case $S^* = 1$ we get the saturation intensity

$$I_{S^*} = \frac{\hbar\omega_0^3}{3\pi c^2} \frac{A}{2} \quad . \quad (\text{A.5})$$

At the TSR the ions are irradiated by linear polarized light, but they themselves are randomly oriented. Therefore we treat the problem as for unpolarized light, where I_s is by a factor of 3 larger. The effect of m_F substates in the $F = 5/2 \rightarrow F' = 7/2$ transition in ${}^7\text{Li}^+$ again reduces I_s by a factor of 2, because we cannot restrict the problem to the $m_F = +5/2 \rightarrow m'_F = +7/2$ case, which is assumed by the condition $\vec{d} \parallel \vec{E}$. Thus the

saturation intensity is

$$I_s = \frac{\hbar\omega_0^3}{2\pi c^2} \frac{A}{2} \quad . \quad (\text{A.6})$$

The numerical value for the above transition in ${}^7\text{Li}^+$ is 8.8 mW/cm^2 .

Appendix B

Measurements with Electron Cooling

Tuning the electron energy to resonance with the ions, a transversely extended ion beam like the ${}^7\text{Li}^+$ beam is initially broadened in momentum space due to the spacially non-homogeneous energy distribution of the electrons across the beam tube [Matl87, Frie89]. The fluorescence linewidth in our case grew almost instantaneously after ion injection to $\delta\nu = 4\text{GHz}$ (see fig. B.1). Due to the long estimated cooling time of 14 s no reduction of the linewidth can be observed.

The radial distribution of kinetic energies in the merged electron beam shows a parabolic dependence. Ions with small betatron amplitudes will thus collide with rather monoenergetic electrons than those moving all across the geometrical acceptance of the TSR. In a Lamb dip measurement similar to that described in section 4.3 with additional resonant electron cooling, it is possible to check the influence on the longitudinal and transverse motion of the ions almost independently. The width of the Lamb dip is determined by the transverse betatron motion, while the total fluorescence linewidth reflects the longitudinal momentum spread of the ion beam. In figure B.1 the widths of the whole fluorescence signal and of the Lamb dip are shown with the cooling time. The total line width is broadened to about 4 GHz after injection and continues broadening due to residual gas scattering (see 4.2). The Lamb dip, however, starts to become narrower after 2 s, much faster than the estimated longitudinal cooling time. This shows that the electron cooling decreases the large transverse motion of an extended ion beam more effectively than reducing the longitudinal momentum spread. In this direction the spatial variation in electron energy counteracts the cooling effect stronger than transversely, where the electrons are magnetically confined.

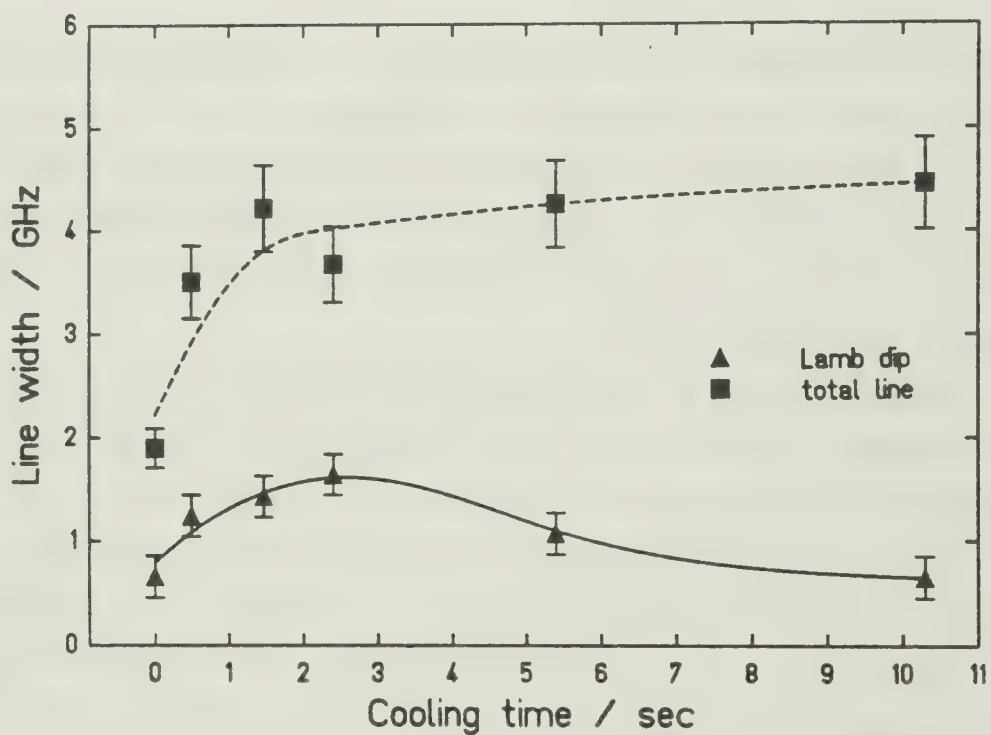


Figure B.1: The total fluorescence linewidth and the width of the Lamb dip are plotted over the cooling time. While the total width constantly broadens, the Lamb dip starts narrowing after 2 s.

Appendix C

Laser Spectroscopy in an Optical Λ -System

In order to prove the feasibility of more complex optical pumping schemes in a storage ring and to show clearly the population changes in the hyperfine sublevels of ${}^7\text{Li}^+({}^3S_1)$, the two laser frequencies are tuned to excite the two transitions in the optical Λ - system shown in figure C.1. The weak standing wave laser is fixed to the $5/2 \rightarrow 5/2$ transition, while the ring dye laser scans the $3/2 \rightarrow 5/2$ transition. In the measurement shown here, the $5/2 - 7/2$ line is scanned first, and the usual Lamb dip is seen. The population transferred to the $F=3/2$ level by the optical pumping is brought to fluorescence, when the Λ -system is closed. The original $F=3/2$ population, which would give a signal of a width similar to the usual fluorescence line of 1.9 GHz (see sec. 4.2), does not contribute here. These ions are optically pumped very quickly, and only those ions can radiate enough fluorescence photons to build up a substantial signal, which are resonant with the fixed frequency laser. Correspondingly the linewidths of the Lamb dip and the $3/2 \rightarrow 5/2$ signal are equal and they match (and are included) into the systematics of figure 4.7. Taking the Doppler shift into account, the measured frequency difference of 7.6 GHz fits with the rest frame transition frequency of 8.04 GHz given in figure 3.3.

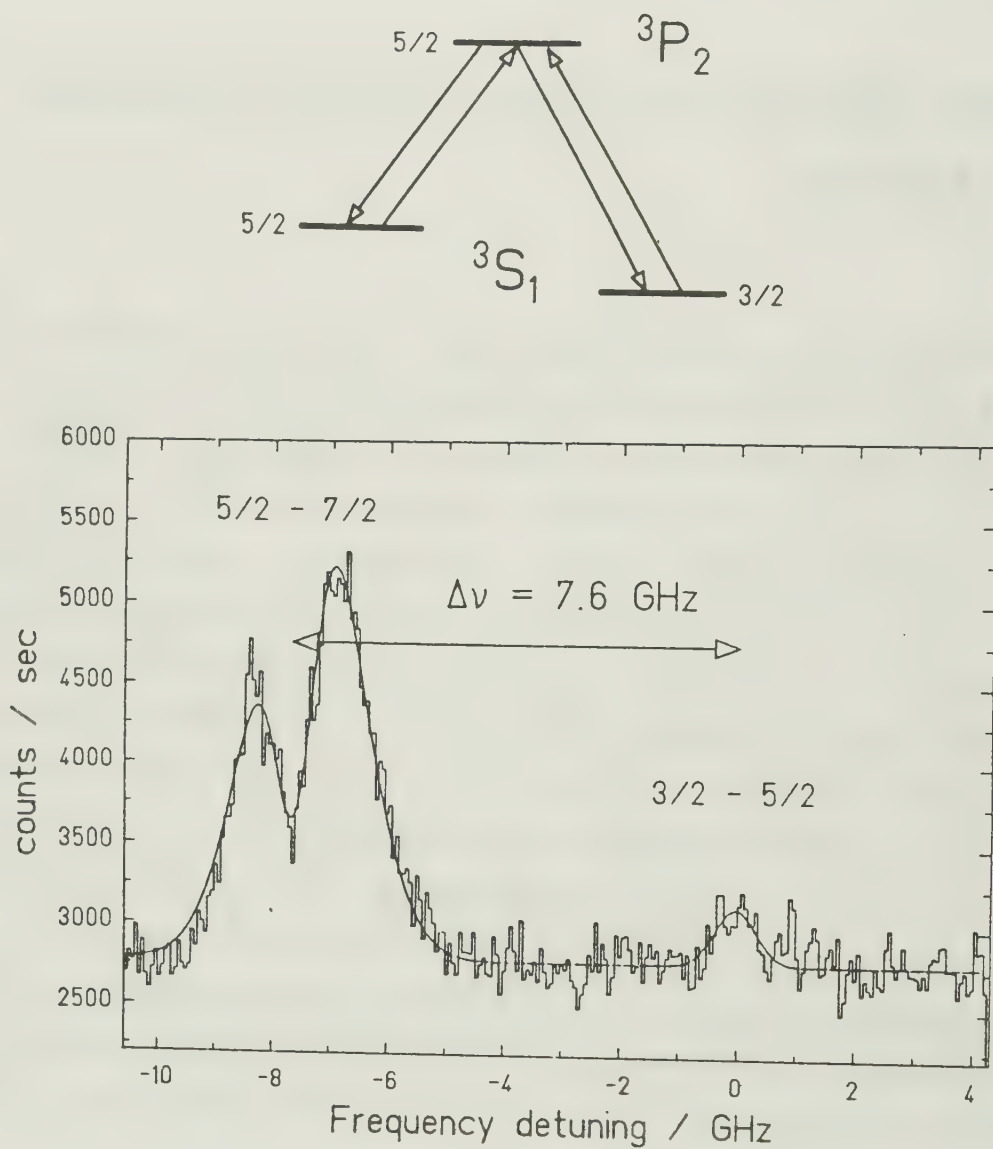


Figure C.1: Fluorescence signal from the optical Λ -system. The laser frequency is scanned across the $5/2-7/2$ and the $3/2-5/2$ transition, while the fixed frequency laser is resonant with the $5/2-5/2$ system. The widths of the $3/2-5/2$ signal is equal to that of the $5/2-7/2$ Lamb dip.

Appendix D

Velocity Scanning Fluorescence Measurements

Velocity scanning measurements similar to those described in section 6.3 are performed in order to measure the lifetime of a laser cooled ion ensemble. When the delay between the laser cooling sweep and the voltage scan is varied, the cooled velocity profile can be probed at different times after the end of cooling. While in figure D.1a both lasers are on during the delay time, the Ar^+ ion laser beam is blocked for the particular voltage scan. From the areas of the corresponding fluorescence signals a storage time of 1.7 s is evaluated. This is twice the value obtained from the fluorescence decay measurement, which, however, is distorted by the velocity mixing effect discussed in section 4.3. Blocking the counterpropagating cooling laser after the laser sweep, yields the series of spectra shown in figure D.1b. Decreasing in intensity with the same time constant of 1.7 s the cooled peak is shifted apart from the Ar^+ ion laser frequency position at zero detuning. This shift stops asymptotically at a detuning of 2.2 GHz, which is connected to the maximum range of the spontaneous light pressure force of the strong Ar^+ ion laser radiation ($S \approx 190$). However, the uncertainties of a voltage scan have to be taken into account. Thus also the increase in line width of a factor of 1.4 or 350 MHz is only of qualitative relevance and should not be interpreted in too great detail.

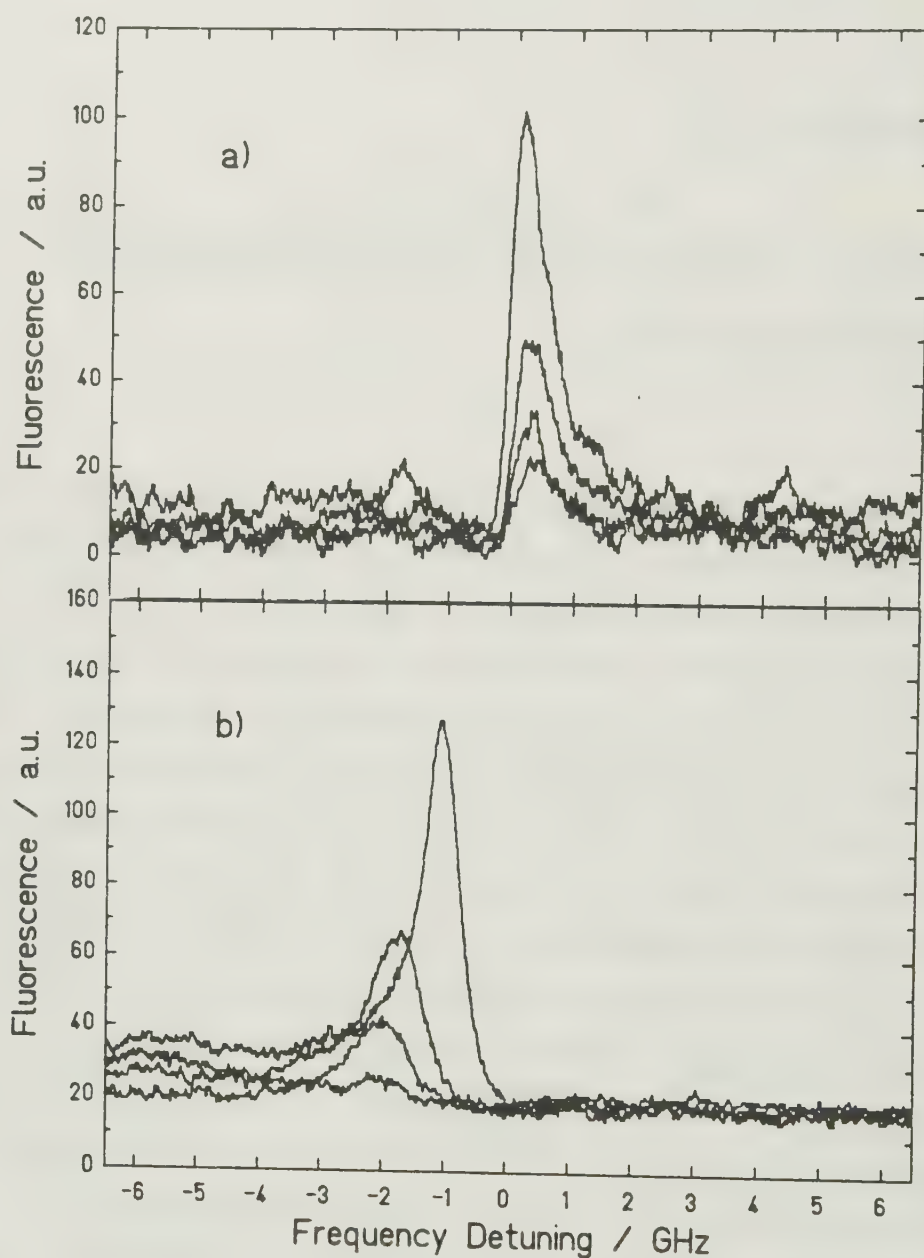


Figure D.1: The cooled velocity distribution is probed after different delays from the end of cooling (0,1,2 and 3 s in (a) and 0,1,2,4 s in (b)). The frequency scale denotes the equivalent detuning of the counterpropagating dye laser. In (a) the ions are irradiated by both lasers for the delay time and by the dye laser during the probing voltage scan, while in (b) the counterpropagating dye laser beam is blocked after the cooling laser scan.

Appendix E

Schottky Noise Signals with Limited Geometrical Beam Control

Before optimization of the geometry by insertion of apertures the laser cooled and shifted ions are detected in the Schottky spectrum in addition to the broad distribution of 1S_0 ground state ions (fig. E.1). The laser cooling sweep is stopped before reaching the Ar^+ ion laser frequency in the ion's rest frame and the RF spectrum analyser is triggered with a delay after ion injection according to the laser sweeping time. The spectra are taken at the 18th harmonic of the revolution frequency and show the revolution frequency distribution of the ion beam without (a) and after laser cooling (b). The deformation of the gaussian shape of the uncooled beam is due to imperfect adjustment of the TSR in this case. The cooled ions are shifted out of the original distribution which is still strongly represented by 1S_0 ions, and to an accuracy, limited by the asymmetric line shape, the amount of momentum change is in agreement with simultaneous fluorescence measurements.

Under the assumption of proportionality of $\int |A(f)|^2 df$ to the number of ions N (see section 6.5) the shifted fraction is about 10% of all ions in the TSR, which is in agreement with the estimate from fluorescence signal intensities (see section 4.2).

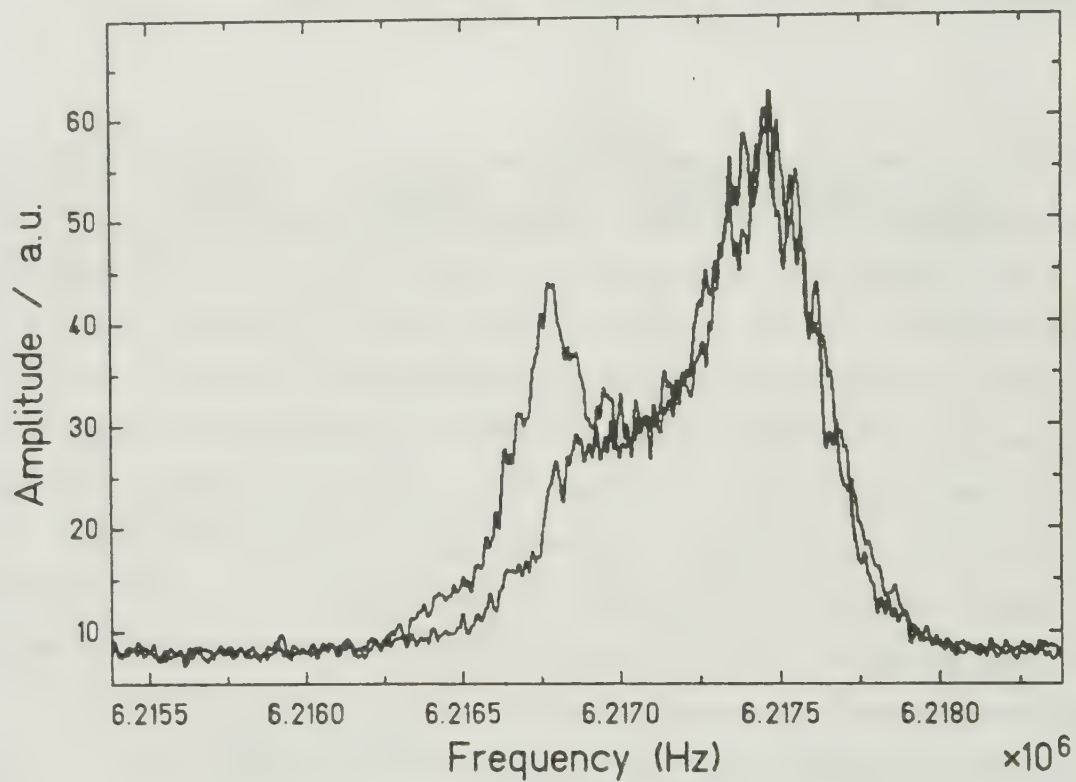


Figure E.1: Schottky noise signals from the stored ${}^7\text{Li}^+$ beam without laser interaction (a) and after laser cooling (b). The spectra are taken at $n=18$ with 3.9 s delay after ion injection.

Appendix F

Noise Suppression in Electron Cooling

In dense ion beams cooled by electron cooling a strong noise suppression is observed and two narrow peaks occur in the Schottky spectrum, which are due to collective waves (sound waves) propagating with and opposite to the ion beam [Poth89, Stec90, Jaes89]. In this case the momentum spread is obviously not directly given by 6.2, but one has to include the detailed theory of plasma waves in a storage ring [Park80, Coch89].

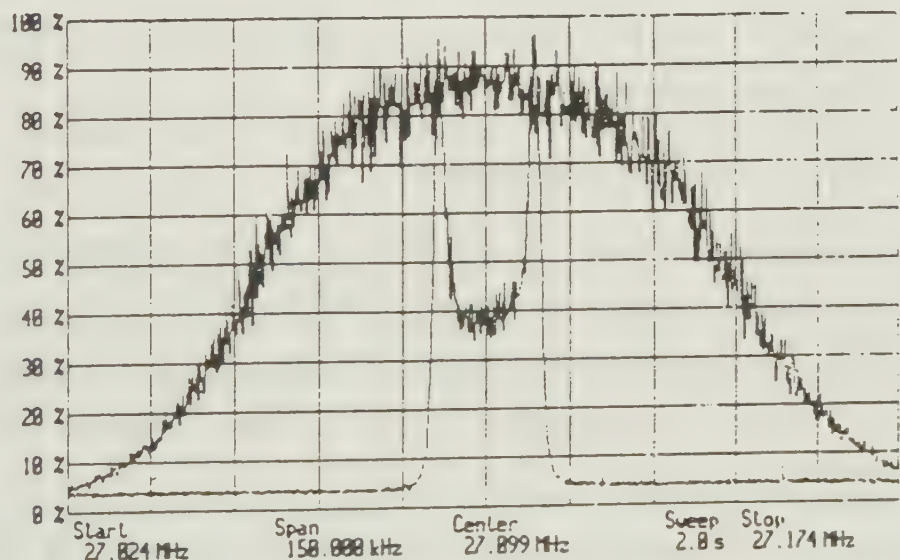


Figure F.1: Schottky spectra of a $^{12}\text{C}^{6+}$ ion beam before and after electron cooling (from [Jaes89]).

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